

Chapter 2B: Mercury Monitoring, Research and Environmental Assessment

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SUMMARY

The problem of mercury in the Everglades has been evident since the late 1980s, when testing of largemouth bass revealed levels of mercury that exceeded all health-based standards. At that time virtually nothing was known about mercury in Florida's environment, including its causes, effects, risks and potential solutions. In response, the Florida Department of Environmental Protection (Department or FDEP) and the South Florida Water Management District (SFWMD or District) organized a multi-agency group, the goal of which would be to work toward understanding the root causes of the mercury problem in Florida. Operating as the South Florida Mercury Science Program, these agencies have improved the predictive understanding of the sources, transformations and fate of mercury in the Everglades. The program has also been effective at linking local information to that at regional and global levels to better support decision making in South Florida and improve the estimation of risks to fish-eating Everglades wildlife. General information on the nature of the environmental mercury cycle has been presented in previous Everglades Consolidated Reports (ECRs) and can be found in Chapter 7 of both the 2000 and 2001 ECR and in Chapter 2B of the 2002 ECR.

Important, updated findings from this collaborative effort on mercury include the following:

- Atmospheric deposition accounts for greater than 95 percent of the external load of mercury to the Everglades. Once deposited, the effect of newly deposited mercury is quickly felt through rapid methylmercury production occurring over a period of hours to days. The relative proportions of local and long-range transport of mercury to the Everglades remain an open question.

¹ This chapter continues the previous year's approach in that it is intended to be accessible to a nontechnical audience. However, **Appendices 2B-1** through **2B-5** provide additional detail to meet the EFA requirement that the District and the FDEP shall annually issue a peer-reviewed report regarding the research and monitoring program that summarizes all data and findings. **Appendix 2B-3** meets the reporting requirements of the Everglades Forever Act, as well as specific permits issued by the FDEP to the District. Readers who desire more detailed scientific information should consult the Chapter 2B appendices in the 2003 ECR, Chapter 7 of the 2000 and 2001 ECRs, Chapter 2B of the 2002 ECR, and related appendices to previous years' ECRs (SFWMD, 1999, 2000, 2001, 2002).

- Methylmercury, the most toxic form of mercury, is produced primarily in sediments. It efficiently bioaccumulates in aquatic food webs and its production is strongly influenced by the rate of supply of atmospherically derived mercury.
- Sulfur follows inorganic mercury deposition rate in order of importance regarding factors controlling methylmercury production and bioaccumulation. Phosphorus plays a secondary role by influencing the carbon, oxygen, and sulfur cycles that affect the methylmercury production rate of sulfate-reducing bacteria.
- The central and southern Everglades exhibit strong methylmercury production and bioaccumulation and, therefore, high mercury levels in fish and wildlife. These levels are high enough to pose a risk of chronic toxicity.
- The primary emissions sources of mercury in Southern Florida *circa* 1990 were incineration (both municipal solid waste and medical waste) and power generation. Mercury emissions from incinerators of all types have declined by approximately 99 percent since the late 1980s. Principal reasons for this decline were reduced uses of mercury and effective emissions controls.
- Monitoring of fish and wading birds indicates a significant decline in mercury by as much as 60 percent in largemouth bass and wading birds at some locations.
- Environmental mercury models for the Everglades have been developed and they incorporate the latest findings from atmospheric and aquatic research. Results substantiate a strong relationship between atmospheric mercury load to the Everglades and mercury levels in top-predator fish.
- Modeling analyses indicate that Everglades response times to changes in atmospheric load are short. Significant benefits could be expected within a decade of load reductions, with ultimate benefits occurring within about 30 years.

The mercury monitoring, research, modeling, and assessment studies described in this chapter and its appendices are coordinated through the multi-agency South Florida Mercury Science Program (SFMSP)² This group of agencies, academic and private research institutions, and the electric power industry have advanced the understanding of the Everglades mercury problem faster and more effectively than either the FDEP or the District could have accomplished individually. The SFMSP has operated under a coordinated plan; however, each agency has operated within its own management and budgeting framework. The goal of the SFMSP studies is to provide the FDEP and the District with information to make mercury-related decisions about the Everglades Construction Project (ECP), as well as other restoration efforts on the schedule required by the Everglades Forever Act (EFA). When this work began, all that was known was that Everglades fish contained unusually high levels of mercury. Now, SFMSP studies are

² This partnership of federal, state and local interests includes the FDEP, the District, the U.S. Environmental Protection Agency (USEPA) Office of Research and Development and Region 4, the Florida Fish and Wildlife Conservation Commission and the U.S. Geological Survey. Other collaborators associated with the SFMSP are the U.S. Fish and Wildlife Service, the U.S. Park Service, the U.S. Army Corps of Engineers, the University of Florida, Florida State University, Florida International University, the University of Miami, the University of Michigan, Texas A & M University, Oak Ridge National Laboratory, the Academy of Natural Sciences of Philadelphia, Florida Power and Light, Florida Electric Power Coordinating Group, the Wisconsin Department of Natural Resources, the Electric Power Research Institute and the National Oceanic and Atmospheric Administration (NOAA).

providing a better understanding of why the Everglades is an “at-risk” system for mercury contamination.

GLOSSARY OF MERCURY-RELATED ACRONYMS AND TERMS

The general glossary in the *2003 Everglades Consolidated Report* (2003 ECR) is designed to support general terminology in all eight chapters. However, because mercury is a complex environmental contaminant involving several specialized scientific disciplines, the following mercury-specific glossary should assist the reader in understanding the material presented in Chapter 2B:

- ACME Project: The Aquatic Cycling of Mercury in the Everglades Project. A process-oriented mercury research program organized by the U.S. Geological Survey.
- E-MCM: Everglades Mercury Cycling Model. A computer model of mercury cycling being refined under the auspices of FDEP, USEPA and SFWMD to predict changes in Everglades mercury in response to changing loads or water quality.
- FAMS: Florida Atmospheric Mercury Study. An early study to quantify deposition of mercury from the atmosphere to the Everglades and other parts of Florida.
- Hg: The standard chemical abbreviation for the element mercury.
- MeHg: The standard chemical abbreviation for methylmercury, a particularly toxic organic form of mercury that concentrates in aquatic food webs.
- REMAP: Regional Environmental Monitoring and Assessment Program. The United States Environmental Protection Agency (USEPA) Region 4, and the Office of Research and Development (ORD) have used the REMAP approach to conduct an Everglades-wide ecosystem assessment for mercury and water quality.
- RGM: Reactive gaseous mercury. A form of gaseous mercury in the atmosphere that is readily deposited by rainfall and dry deposition.
- SFMSP: South Florida Mercury Science Program. A state-federal-private partnership to determine the causes of and solutions to the mercury problem in Florida.
- SRB: Sulfate-reducing bacteria. Microbes, commonly found in sediments, that transform inorganic mercury into methylmercury.
- TMDL: Total Maximum Daily Load. Load determinations for a waterbody not meeting its designated use as required under the Clean Water Act.

RESEARCH PROGRESS

The following research needs were identified in the 2000, 2001 and 2002 Everglades Consolidated Reports. A brief update on research progress on each need is presented below.

1. Quantify the wading bird diet-egg relationship to support a revised numerical Class III water quality standard for total mercury based on methylmercury levels (2000 ECR). Ecological Risks of Mercury (2001 ECR).

The U.S. Department of the Interior (DOI) U.S. Fish and Wildlife Service (USFWS) Patuxent Wildlife Research Center initiated a study of the *in-ovo* effects of methylmercury. Dr Gary Heinz, principal author of the much-cited study of the multi-generational effects of mercury on domestic ducks, has obtained extensive collections of the fertile eggs of several wading bird species and has begun detailed studies of egg viability and hatchability. District staff continue to provide in-kind support for Dr. Heinz's research by collecting and providing eggs gathered from South Florida nests.

2. Quantify global versus local, and new versus old sources of mercury (2001 ECR). Local source: Receptor Relationships of Mercury (2002 ECR).

The Department and the USEPA continue to support atmospheric mercury studies relevant to mercury control policy in southeast coastal regions. The agencies presently sponsor studies that directly measure transport of mercury species into Florida, describe and quantify the atmospheric reactions of mercury that lead to deposition, and develop models to organize the atmospheric processes research into decision-making tools. Continuing since the previous Everglades Consolidated Report, the Department and the Broward County Department of Planning and Environmental Protection, Air Quality Division, operate two intensive air-monitoring sites in Broward County. This project, the Speciated Atmospheric Mercury Study (SAMS), recognizes the paramount importance of the role played by speciation of mercury in the atmosphere in controlling the transport and fate of mercury. SAMS makes highly time-resolved measurements of all known forms of atmospheric mercury and associated tracer species. This measurement and modeling project will continue through 2003 and will provide improved data and tools and will facilitate understanding of the question of the importance of long-distance transport of mercury into Florida.

Fieldwork on these interrelated projects has proceeded apace, but sample analysis provided at no cost by the USEPA has been slowed by equipment failure. A one-of-a-kind analytical instrument – a high-volume, automated energy-dispersive x-ray fluorescence (EDXRF) spectrometer developed for the USEPA by Lawrence Berkeley Laboratories (LBL) in the 1970s – began exhibiting degraded performance in 2000. This instrument was returned to LBL for repair and upgrade and was subsequently returned to the USEPA in late spring 2002. This instrument is essential in measuring the multi-element suite of elemental tracers (30 to 40 elements, ranging from aluminum to zinc) that allow for estimation of the origin, characteristics and source influences of air masses sampled. The EDXRF has been tested and calibrated and is in routine operation. Analysis of the backlog of hundreds of Florida samples should be completed by the end of 2002.

3. Revise the Everglades Mercury Cycling Model (E-MCM) to include food web uptake dynamics and relationships between phosphorus and sulfur concentrations and mercury dynamics (2001 ECR).

Research that will define the details of the mercury methylation process and the quantitative relationships with the factors that influence it is key to learning the factors that

control the production of MeHg in the aquatic system. This will be the SFMSP's focus during 2001 through 2003. As this work progresses, the information gained will be incorporated into the evolving E-MCM to make it a more robust tool for evaluating management options. Data and insight from field studies will be fed directly into model formulation and testing. The results of this work will be a calibrated and tested aquatic mercury cycling model that can simulate the effects of various hydrology, water quality, or restoration activities.

A spatially explicit version of the E-MCM, designated E-MCM2, is under development to assess system-wide responses to mercury sources, water quality, and management scenarios being evaluated by the Comprehensive Everglades Restoration Plan (CERP).

4. Geochemical controls on mercury methylation (2001 ECR).

The FDEP continues to fund a series of studies with the U.S. Geological Survey and the Academy of Natural Sciences of Philadelphia, Estuarine Research Laboratory. A detailed update on these projects is given in Chapter 2B-2 of the 2002 ECR. These studies use field mesocosm experiments, with stable-isotope and other tracer techniques, to examine the interactions between mercury, sulfur, nutrients and other water quality variables. Fieldwork began with deployment of mesocosms in spring 2001; field experiments are scheduled through June 2005.

5. Trends of mercury in Florida (2002 ECR).

Evaluating mercury trends over time can be one of the most illuminating uses of the data. For example, the sediment coring studies of the early 1990s by Rood et al. (1995) revealed that mercury accumulation in Everglades soils was more than five times greater than in 1900, confirming the view that anthropogenic influences dominated mercury cycling. Following that study, there is a hiatus of 10 critical years in the record to describe the direction and magnitudes of mercury impinging on South Florida. To close that gap in the knowledge, the FDEP is revisiting that work by selecting three to five waterbodies that have sedimentary profiles more ideal than those in the Everglades. A team of scientists from the Science Museum of Minnesota, the University of Florida, the University of Connecticut, and Tetra Tech, Inc. will complete the fieldwork in 2002. The results of this project should yield high-resolution information on the trend of mercury accumulation in South Florida. These data, in conjunction with other trend information developed by the FDEP and its collaborators will allow evaluation of the outcomes and effectiveness of controls on mercury use and emissions.

In late 2001 and early 2002, multiple cores were collected at each of three lakes in South Florida. Preliminary analyses to date indicated that sediment profiles for these lakes are suitable for dating, and the lead-210 dating is underway. Initial results are due by June 2003. The group of investigators was awarded a USEPA STAR grant for similar studies, some of which will be coordinated with the STAR-funded projects. Following the initial phase funded by the FDEP, coring of additional lakes is planned for 2003, with project completion scheduled for mid-2004.

KEY FINDINGS ON THE EFFECT OF WATER QUANTITY AND QUALITY ON METHYLMERCURY PRODUCTION IN REPORTING YEAR 2003

From a management perspective, no subject in mercury cycling is more important than the factors involved in transforming inorganic mercury into methylmercury. A major emphasis has been placed on careful examination of methylmercury production. Research in this area is summarized in **Appendix 2B-2**. Key findings are provided below:

- At sites immediately downstream of South Florida Water Management District structures, the loading per unit area (flux) of inorganic mercury and methylmercury from Lake Okeechobee releases and Everglades Agricultural Area (EAA) runoff is greater than that from wet and dry atmospheric deposition. This suggests that removal of these species by the Stormwater Treatment Areas (STAs) could have a beneficial impact on the northern Everglades.
- Methylmercury is synthesized primarily from both new inorganic mercury being supplied by runoff and from wet and dry atmospheric deposition, not from soil release, even following a dryout event.
- Methylmercury production is much more temperature sensitive than methylmercury decomposition; this temperature sensitivity changes with location, suggesting that different microbial communities are involved in methylation in these locations.
- Once methylmercury is synthesized in surficial peat soils, transport from soil to water occurs, which is probably mediated by daily movements of microorganisms and macroorganisms living on and in the soil.
- Once methylmercury is present in the water column, decomposition by sunlight (photodegradation) competes with sorption to settling organic particles as the most significant removal pathway from the water column.
- The concentrations of sulfide in pore water and soil influence methylmercury production by increasing the bioavailability of inorganic mercury to methylating bacteria. Whether the uptake of the inorganic mercury-sulfide complex occurs through passive (diffusion) or active (facilitated) transport remains open to debate. In contrast, excess sulfide can reduce the availability for methylation.
- The addition of sodium sulfate, sodium sulfide and a slurry of ferrous sulfide (pyrite) inhibited net methylmercury production in laboratory microcosms.
- Iron appears to play a role in mediating sulfur and mercury speciation, and stimulation of methylmercury production at Water Conservation Area (WCA) 3A-15 was observed when ferrous chloride was added to a soil slurry. The mechanism(s) by which this occurs have yet to be elucidated.

THE MULTIMEDIA CYCLE OF MERCURY

The accumulation of mercury in fish is one of the Everglades water quality problems the District and the FDEP are addressing in their activities under the Everglades Forever Act (EFA) and, in the future, the Comprehensive Everglades Restoration Plan (CERP). The mercury problem first became apparent in 1989, when the Florida Department of Health issued mercury-related health advisories to fishermen. These recommendations, the first ever in Florida, urged fishermen not to eat largemouth bass, a popular sport fish, from most of the Everglades, and to consume only limited amounts of several other species of sport fish because of a risk of mercury toxicity. The high levels of mercury found in fish also pose a risk of mercury toxicity to fish-eating wildlife.

The conceptual view of the mercury problem in the Everglades, as in many other waters, is that it is a multi-media problem, meaning that more than one aspect of the environment is involved. To understand the problem's individual components, one must combine the disciplines

of air quality with water quality and ecological risk. The conceptual model of the mercury problem is shown in **Figure 2B-1**.

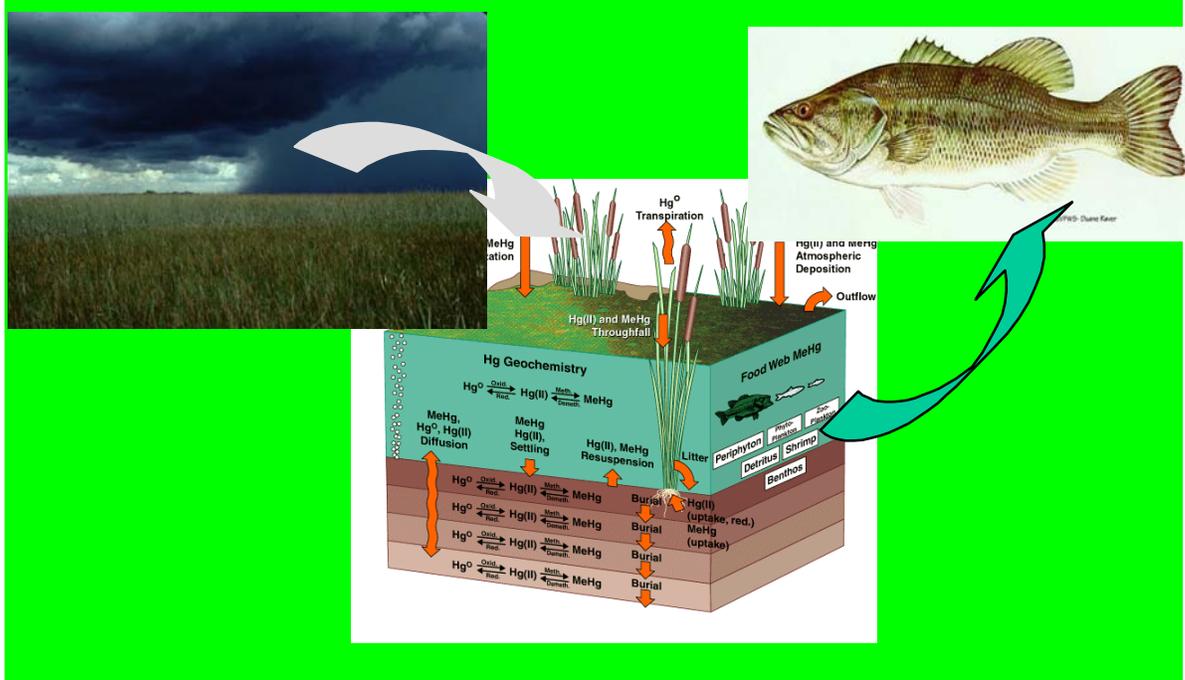


Figure 2B-1. The multimedia aspects of the Everglades mercury problem, encompassing sources from air deposition, biotransformation in the aquatic system and bioaccumulation through the aquatic food web. Photo by D. Scheidt, USEPA; Graphic by Tetra Tech

The most important concepts to keep in mind regarding the environmental mercury cycle are as follows:

- Air Pollution and Deposition.** Since the Industrial Revolution, human mining and industrial activities have increased the amount of mercury that naturally cycled through the atmosphere by about five-fold. Mercury pollution in the air comes from the mining and smelting of mineral ores (which contain small amounts of mercury), the burning of fossil fuels, such as coal and oil, the use and disposal of mercury, and the incineration of wastes, principally municipal and medical wastes. The predominant source of mercury to the Everglades is from atmospheric deposition. Mercury deposited by rainfall is about 50 times greater than the amounts of mercury in the discharges of surface water to the Everglades. Dry deposition of gaseous and fine particulate mercury adds another one-third of rainfall deposition. Though the ultimate source of mercury is from the air, amounts of mercury in lakes, rivers and Everglades waters are not high enough to be directly harmful to humans, fish or wildlife.
- Biotransformation and Bioaccumulation.** Once deposited to the Everglades, mercury is quickly distributed through the shallow water column and into sediments, where a

fraction, typically a few percent, is transformed by naturally occurring bacteria to methylmercury. Unfortunately, methylmercury is very toxic and it also bioaccumulates efficiently from the water up through aquatic food webs.

- **Ecological Risk.** Methylmercury in the Everglades top-predator fish (such as largemouth bass) has been measured to be as much as 10 million-fold higher than that of the water in which the fish swim. This phenomenal bioaccumulation of mercury results in a risk of mercury toxicity to animals, including humans, wading birds and other animals that feed on fish.

In departure from its predecessors, this chapter is organized around these three key concepts that describe this problem.

THE ATMOSPHERIC MERCURY CYCLE

When mercury was first discovered in Everglades fish in the late 1980s, little was known of the causes of the mercury problem. Answers to even the most basic questions regarding mercury, such as whether the presence of mercury was simply a natural Everglades condition, whether it had always been that way, source(s) of the mercury, how much was safe or harmful, and most importantly what could be done to reduce or alleviate mercury levels, were unknown.

These conundrums were considered by the Mercury in Fish and Wildlife Task Force, created by the governor in late 1989. The task force ultimately approved the preparation of a report calling for a broad range of environmental studies that would attempt to find answers to mercury-related questions. At that time there were few precedents for the concept that air quality could exert a significant influence on surface water quality. However, in 1992 the FDEP, the District and the USEPA prepared a comprehensive study plan that called for evaluation of both air and watershed sources of mercury to the Everglades.

Table 2B-1 and **Figure 2B-3** provide the results of two years of monitoring comparing atmospheric mercury deposition to Florida (by the Florida Atmospheric Mercury Study, FAMS), with biweekly monitoring of surface water discharges to the Everglades Protection Area (EPA) via the “into” structures discharging to the EPA. These data revealed that atmospheric deposition of mercury is about 50 times greater than surface water inputs (Stober et al., 1996; 1998; 2001, Pollman et al., 1995; Landing et al., 1995; Guentzel, et al., 1998, 2001).

Year	Atmospheric Deposition kg/yr ³	EAA Water Discharge Hg kg/yr ⁴
1994	238	2
1995	206	3 to 4

³ Annual mercury deposition derived from Florida Atmospheric Mercury Study (FAMS), 1993 to 1997.

⁴ Derived from biweekly monitoring of “into” structures discharging from the EAA into the Everglades Protection Area, USEPA, 1994 to 1995 (Stober, *et al.*, 1996).

A complementary study of mercury accumulation in dated sediment cores from the Water Conservation Areas (WCAs) and Everglades National Park (Park or ENP) revealed that the rate of mercury accumulation in Everglades soils at the top of the cores, i.e., circa 1992, was approximately six times higher than the core strata representing 1900. (Rood, et al., 1995). The mercury study altered the perception of the Everglades as a pristine resource and fostered the view that the Everglades was contaminated by external influences. The Everglades could no longer be viewed as unimpacted by pollution. At that time, neither the sources of mercury nor the transport systems that were delivering it were evident (**Figure 2B-2**).

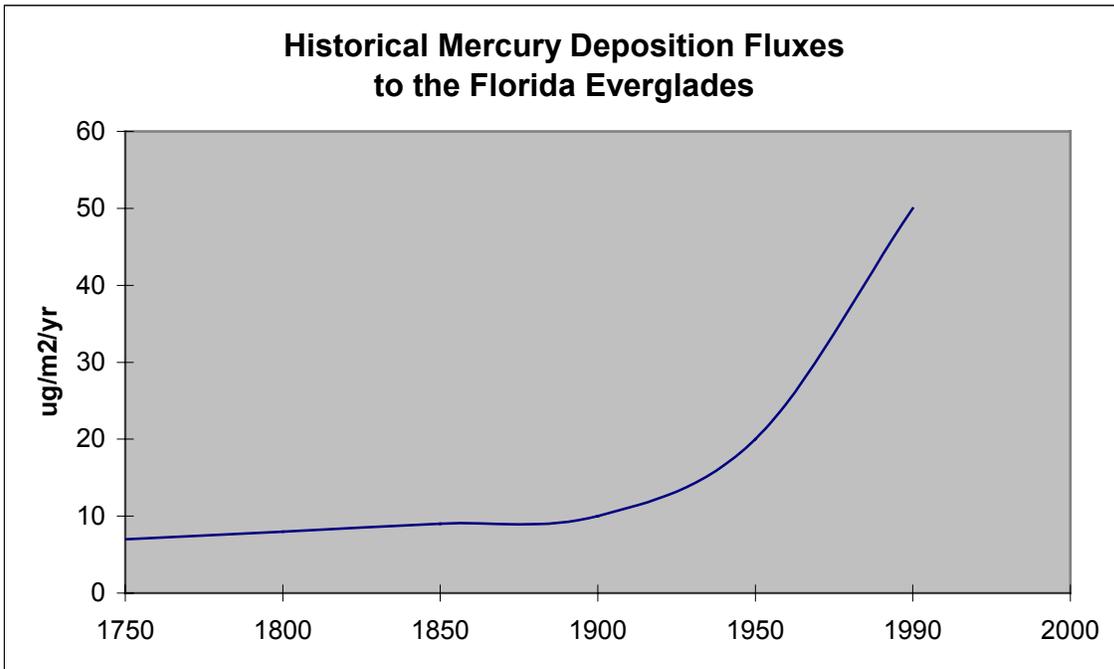


Figure 2B-2. Generalized depiction of long-term trend of accumulation of mercury in soils, broadly applicable to the Everglades and many other waterbodies

Despite the remaining uncertainties regarding the sources and routes of mercury contamination in the Everglades, the two studies illustrated above crystallized the perception that in terms of its mercury budget, the Everglades is a system contaminated by anthropogenic activities principally mediated by atmospheric transport and deposition.

Because atmospheric deposition of mercury is the dominant source of mercury to the Everglades, the FDEP has pursued pollution prevention and emissions controls as having the greatest likelihood for controlling the mercury problem. Major reductions in mercury use and emissions in southern Florida have been achieved, thus, it is hoped, decreasing the delivery of atmospheric mercury to the Everglades. Findings from both environmental monitoring and computer models suggest control of atmospheric sources of mercury can have positive benefits for the Everglades Protection Area. Elimination of mercury from commercial and industrial products and processes since the late 1980s has reduced mercury emissions from municipal waste incinerators and other sources in South Florida.

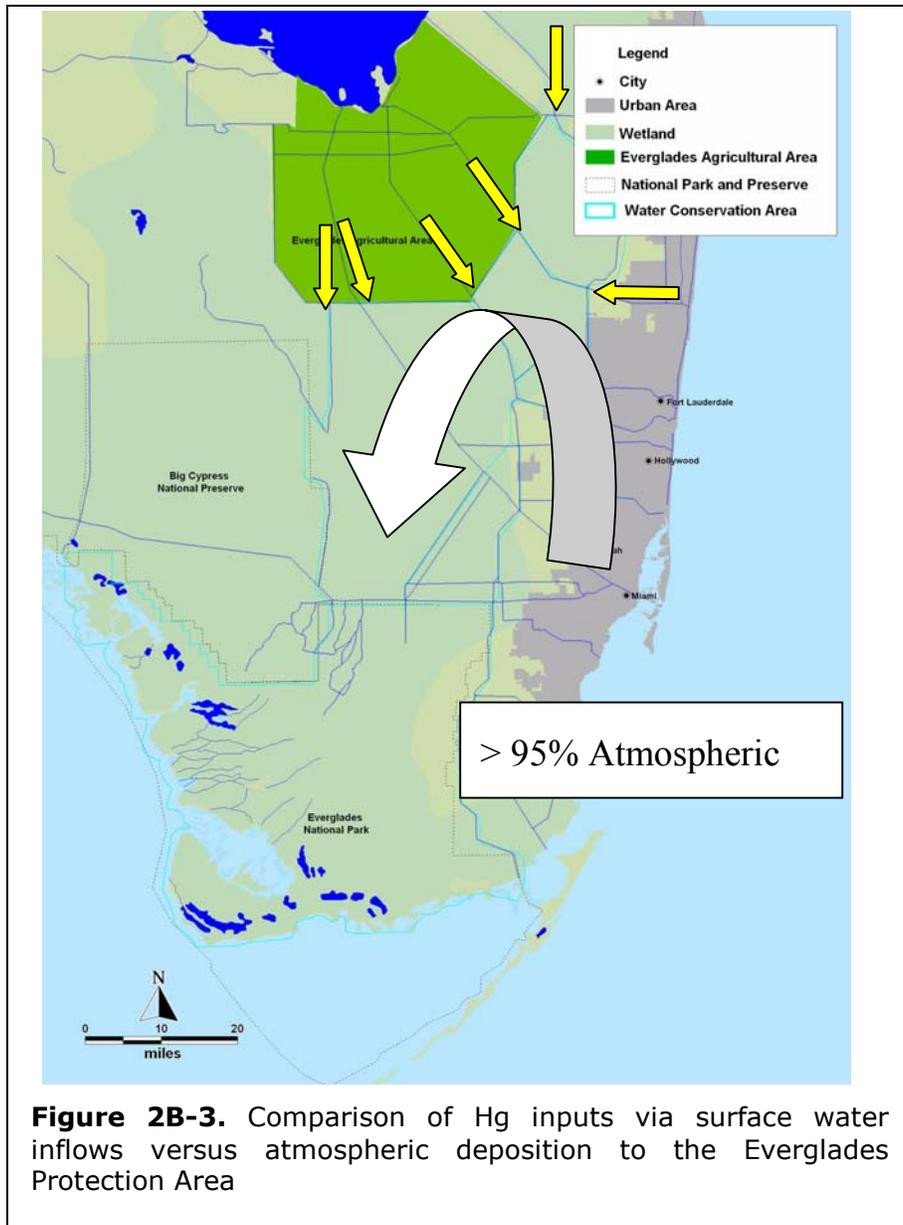


Figure 2B-3. Comparison of Hg inputs via surface water inflows versus atmospheric deposition to the Everglades Protection Area

Monitoring over the past decade suggests that these lower emissions have produced a corresponding reduction in mercury burdens of Everglades fish and wading birds. Environmental models developed by the Mercury Science Program relate fish mercury levels to the amount impinging on the Everglades. These models show that control of mercury emissions should significantly alleviate the overall Everglades mercury problem within a decade or two.

If control of local emissions was not sufficient, it might be possible to reduce the mercury problem through management of water quality and quantity. This approach would make environmental conditions less favorable for the production of methylmercury. Management of marsh fire frequency, hydrologic patterns and water constituents, such as sulfate, may provide a means for such mitigation. With either approach, less methylmercury would be available, making the accumulation of toxic amounts in fish and wildlife less likely.

Another important policy question about atmospheric mercury is its sources. The primary transport regimes relevant to South Florida are local scale (i.e., transport times of approximately one day, or ca. 100km) and global scale (i.e., transport times of weeks, dispersing over much of the globe) (Expert Panel, 1994). Florida is largely disjunct from the regional background of emissions that dominates the North American interior. This analysis suggests that local scale sources are the most important contributors of mercury to the Everglades, as shown in **Figure 2B-4**.

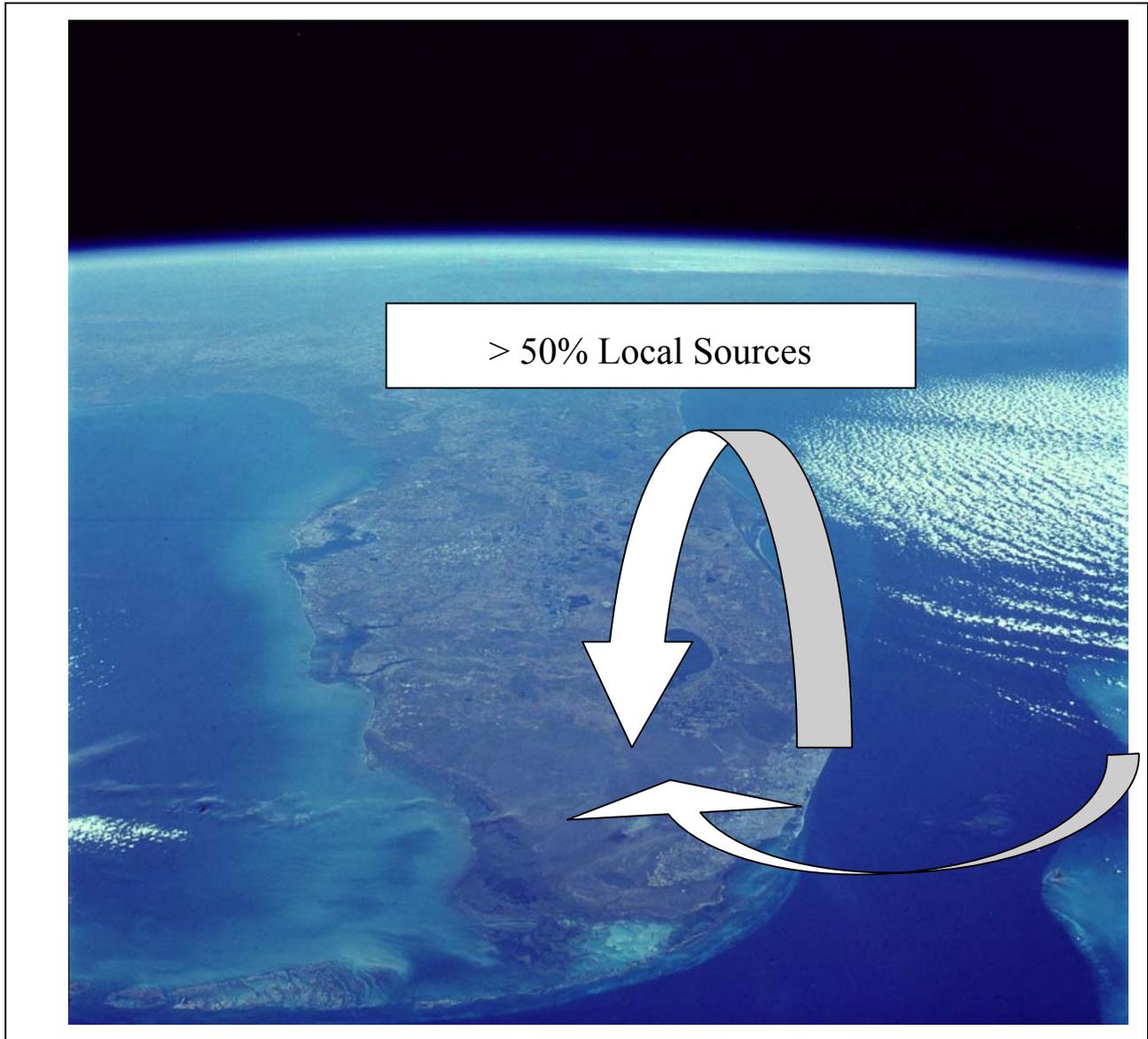


Figure 2B-4. Importance of locally derived mercury to deposition into the Everglades

Weight of Evidence Analysis of Mercury Trends

As is illustrated in **Figure 2B-4**, and in recognition that there is only limited information available to support modeling of a global transport domain, source-receptor modeling relied primarily on local sources to estimate mercury deposition to the Everglades. This has led to considerable controversy over the contributing role of local emissions in South Florida to wet deposition rates in the Everglades, and has fueled debate that the significance of the global or “long distance transport” source to the total deposition signal is too uncertain to conclude whether

local sources are important. It is possible, however, to use several lines of evidence available to bound the potential contributions of mercury in Florida:

- Using multivariate receptor modeling, Dvonch, et al. (1998) concluded that 71 ± 8 percent of the wet deposition signal measured at five sites in the Everglades could be accounted for by local sources. Conversely, Guentzel, et al. (2001), based on their analysis of seasonal patterns in wet deposition of mercury and the uniform nature of summertime mercury concentrations in rain across the South Florida, as well as source apportionment calculations based on a relatively simple mass balance box model on atmospheric fluxes of reactive gaseous mercury in South Florida, concluded that local sources can account for only 30 to 46 percent of the wet deposition signal.
- An Everglades-wide sediment coring study begun in 1992 (Rood, et al., 1995) yielded estimates of historical mercury accumulation rates in Everglades soils spanning the period 1900 through ca. 1990. Comparison with recent deposition estimates (as $\mu\text{g}/\text{m}^2/\text{yr}$) among comparable Everglades sites from that time to the present are given in the table below:

Rood, et al., 1995	ca. 1990	NADP MDN 2002	2001
WCA-1	79	ENR Project	21
WCA-2	59	NA	--
WCA-3	39	Andytown	24
ENP	40	ENP	18

The average mercury accumulation rate, Everglades-wide, in sediment was $53 \mu\text{g}/\text{m}^2/\text{yr}$ ca. 1990 versus $21 \mu\text{g}/\text{m}^2/\text{yr}$ from atmospheric deposition in 2001. These data suggest a decline in deposition of ~ 60 percent overall since ca. 1990.

- Everglades largemouth bass (fillet) and great egret (feather) mercury concentrations have declined ca. 60 percent from the mid-1990s to 2002.
- Data on potential trends in the “global background” indicate a “very small downward trend from 1995 through 2000” at Alert, Canada (Schroeder, Pers. Comm., 2002) and no trend at Mace Head, Ireland (Ebinghaus, et al., 2002). Both are relatively remote background sites. Published data indicate a decline over the northern Atlantic of ca. 20 percent by the mid-1990s. (Langer and Slemr, 1991; Slemr, et al., 1995; Slemr, 1996; Slemr and Scheel, 1998; Ebinghaus and Slemr, 2000). It is apparent that long-term trends of elemental mercury in the atmosphere at background sites are not of similar magnitude to the declines evident in the Everglades ecosystem.
- An independent analysis by the Florida Electric Power Coordinating Group on trends of mercury emissions and concentrations in South Florida biota (per above) concluded that at this juncture, “. . .it is clear that the fundamental hypothesis that changes in local emissions of mercury [in southeast Florida] have been the primary agent for recent biota changes in mercury concentrations in the Everglades cannot be rejected.” (Pollman and Porcella, 2002).
- Receptor modeling applied to source and ambient data during the intensive SoFAMMS field study in 1995 indicated that $92 (\pm 30)$ percent of the total mercury deposition measured at the Davie site near Ft. Lauderdale could be accounted for by local sources (Dvonch, et al., 1998).
- The approximate magnitude of the global background contribution can be bounded from the ca. 300 rain samples from 17 sites in Dade and Broward counties during the August 1995 SoFAMMS project. Observed minima in background rainfall mercury concentrations at

coastal sites in South Florida approximated 5 ng/L. Assuming annual rainfall rates of 130 cm/yr., these concentrations would result in background deposition rates of about 6.5 $\mu\text{g}/\text{m}^2/\text{yr}$., or about 21 percent of rainfall deposition.

- As reported herein, comparative model analyses of transport of mercury from all point sources in Florida and adjoining states versus the 38 point sources in the South Florida modeling domain alone indicated a contribution to the Everglades from regional sources within the southeastern United States of less than 5 percent of total deposition.
- In contrast, the FAMS investigators (Landing, et al., 1995, Guentzel, et al., 1997, 2001) concluded from the weak trace element signatures in rainfall samples and a box model of mercury fluxes into and over Florida that local sources alone could not account for the amounts of mercury measured in rain.

The FDEP views the FAMS and SoFAMMS projects as complementary, not contradictory. Combined with other information, both projects support the notion that the dominant source term signal contributing to total mercury deposition in South Florida is local emissions.

By FDEP analysis, the estimated total deposition for June 1995 through June 1996 was 35.3 $\mu\text{g}/\text{m}^2/\text{yr}$ (Atkeson, et al., in prep.), of which 23 $\mu\text{g}/\text{m}^2/\text{yr}$ was measured by FAMS as wet deposition, and 12.2 $\mu\text{g}/\text{m}^2/\text{yr}$ was modeled as derived from dry deposition. Dry deposition in South Florida expectedly is greatly dominated by RGM. Since the removal rate of RGM from the lower troposphere is rapid, and because the production rate is low, it is reasonable to assume that the predominant fraction of the dry deposited mercury in the Everglades is local in origin. If we assume that this fraction is 80 percent, then this equates to a local contribution of 9.8 $\mu\text{g}/\text{m}^2/\text{yr}$. We then take the lowest estimate of 30 percent from Guentzel, et al. (2001) to describe the local emissions contribution to the annual wet deposition of 23.12 $\mu\text{g}/\text{m}^2/\text{yr}$. This equates to a lower limit contribution of 6.9 $\mu\text{g}/\text{m}^2/\text{yr}$ from local emissions.

Combined, the total estimated contribution from local emissions to wet and dry mercury deposition is 16.7 $\mu\text{g}/\text{m}^2/\text{yr}$, or 47 percent of the total signal. If a contribution of 6.5 $\mu\text{g}/\text{m}^2/\text{yr}$ is ascribed to the global background (as described above), then the maximum that other regional and larger-scale sources other than global background can contribute is 34 percent. If the midpoint of the Guentzel, et al. (2001) estimate of local contributions to wet deposition (38 percent) is used, then the contributions of each major source category to total deposition are as follows:

- Local sources: 52.5%
- Global background: 18.4%
- Other regional sources: 29.1%

Regardless of whether the FAMS or SoFAMMS analyses discussed previously ultimately prove to be closer to the truth, their combined results can be used to constrain a lower limit for the likely contribution of local sources to total deposition. It is the FDEP's conclusion that the sum of these various lines of argument suggests that at a minimum, local sources account for more than 50 percent of mercury deposited in South Florida; several other analyses suggest the contribution may be substantially greater. Narrowing of these divergent estimates is one of the remaining goals of the SFMSP.

COMPREHENSIVE SOURCE REDUCTION

Finding remedies for the problem of excessive mercury in fish has been limited by predictive knowledge of its causes. However, one general aspect of the solution is clear: mercury emissions to the environment should be limited where available information and technology allow. The FDEP has vigorously pursued the following approaches:

- **Pollution Prevention.** The 1993 Florida Solid Waste Management Act required the elimination of mercury from some commercial products and a reduction of the mercury content in wastes. The act bans the use of mercury in packaging materials, prohibits incineration of mercury-containing devices, promotes recycling of such products, and phases out the use of mercury-containing batteries. Presently, international treaties within North America and between North America and Europe are seeking further reductions in the use of mercury.
- **Waste Disposal.** Hazardous waste regulations have been tightened to require stricter control of mercury-containing wastes. Proper disposal minimizes long-term releases of mercury into the environment. A side effect of stricter regulation of mercury discharges has been to encourage elimination of mercury from commercial products and industrial processes.
- **Emissions Control.** A Florida emissions inventory found that the major sources of atmospheric mercury were municipal solid waste combustors, medical waste incinerators and electric utility boilers. The FDEP adopted the first U.S. regulations limiting emissions of mercury from waste combustors, and has also adopted USEPA regulations for medical waste incinerators. Solid waste combustor emissions controls are in place on most Florida facilities in, and MWI emissions have dropped sharply as the industry has moved away from incineration in response to emissions regulations. Emissions in Florida from each of these sectors have dropped more than 90 percent since 1990.

As is shown in **Figure 2B-5** and **Figure 2B-6**, these actions have resulted in significant declines in the use of mercury in commercial and industrial applications, and corresponding reductions in mercury emissions to the South Florida atmosphere.

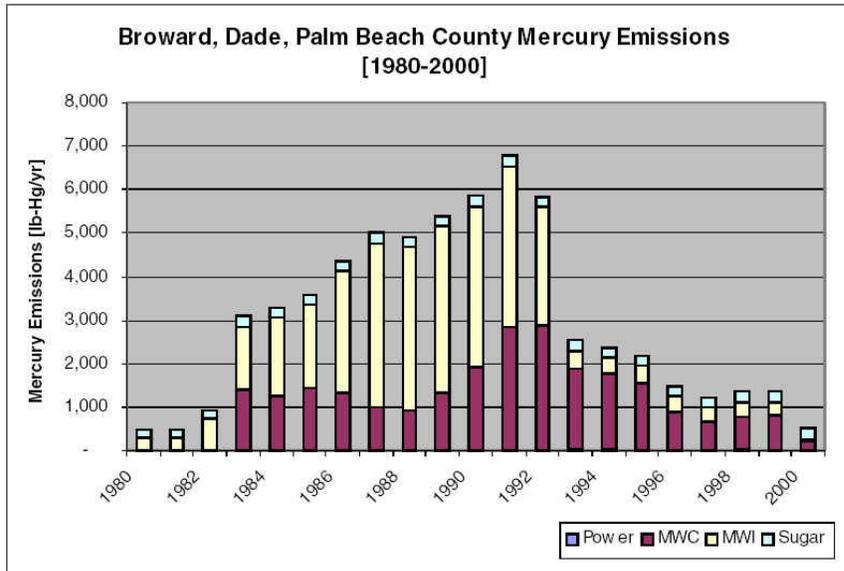


Figure 2B-5. Reconstruction of industrial mercury emissions in Dade, Broward and Palm Beach counties, Florida. Abbreviations: MWC = municipal waste combustors; MWI = medical waste incinerators. RMB Consulting and Associates, 2002

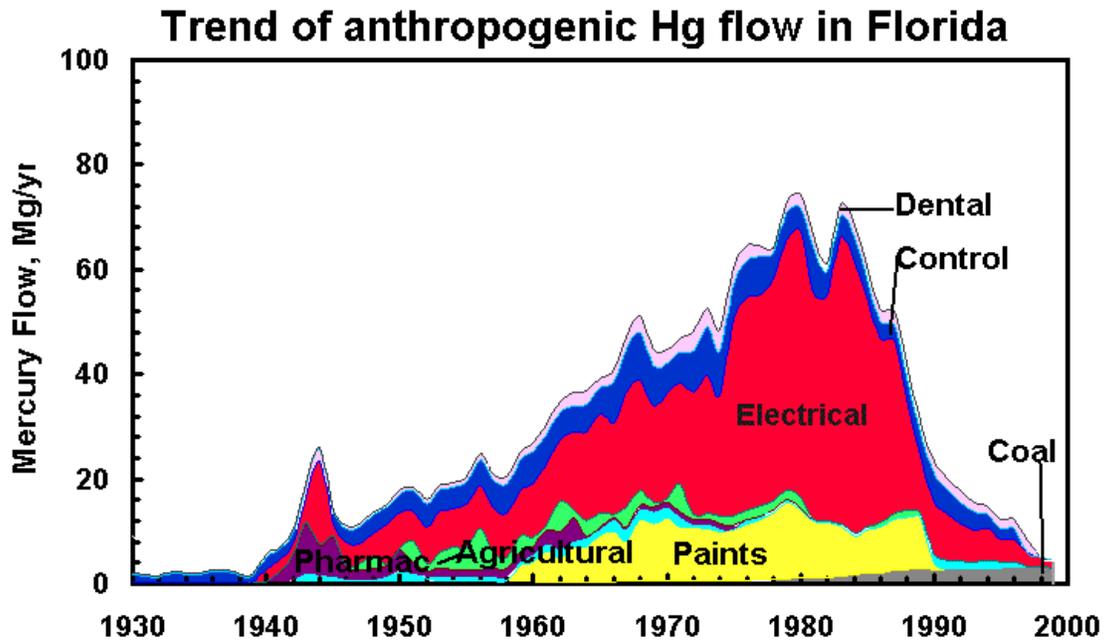


Figure 2B-6. Trends in mercury use in Miami-Dade, Broward and Palm Beach counties, Florida

Because the first full year of atmospheric mercury deposition monitoring in South Florida did not take place until 1994, the FDEP has until recently been unable to determine whether mercury emissions controls have had an influence. Although more extensive work is ongoing, the most useful information shows the success of various control policies (**Figure 2B-7**). FDEP and USEPA initiatives have resulted in reduced uses of mercury in South Florida (i.e., pollution prevention) and emission reductions (e.g., passage of the FDEP MSW incinerator rule in 1994 and full implementation of the USEPA Maximum Achievable Control Technology [MACT] regulations in 2000) that appear to have resulted in significant declines in the atmospheric deposition of mercury to the Everglades and South Florida (Robbins, et al., unpublished data).

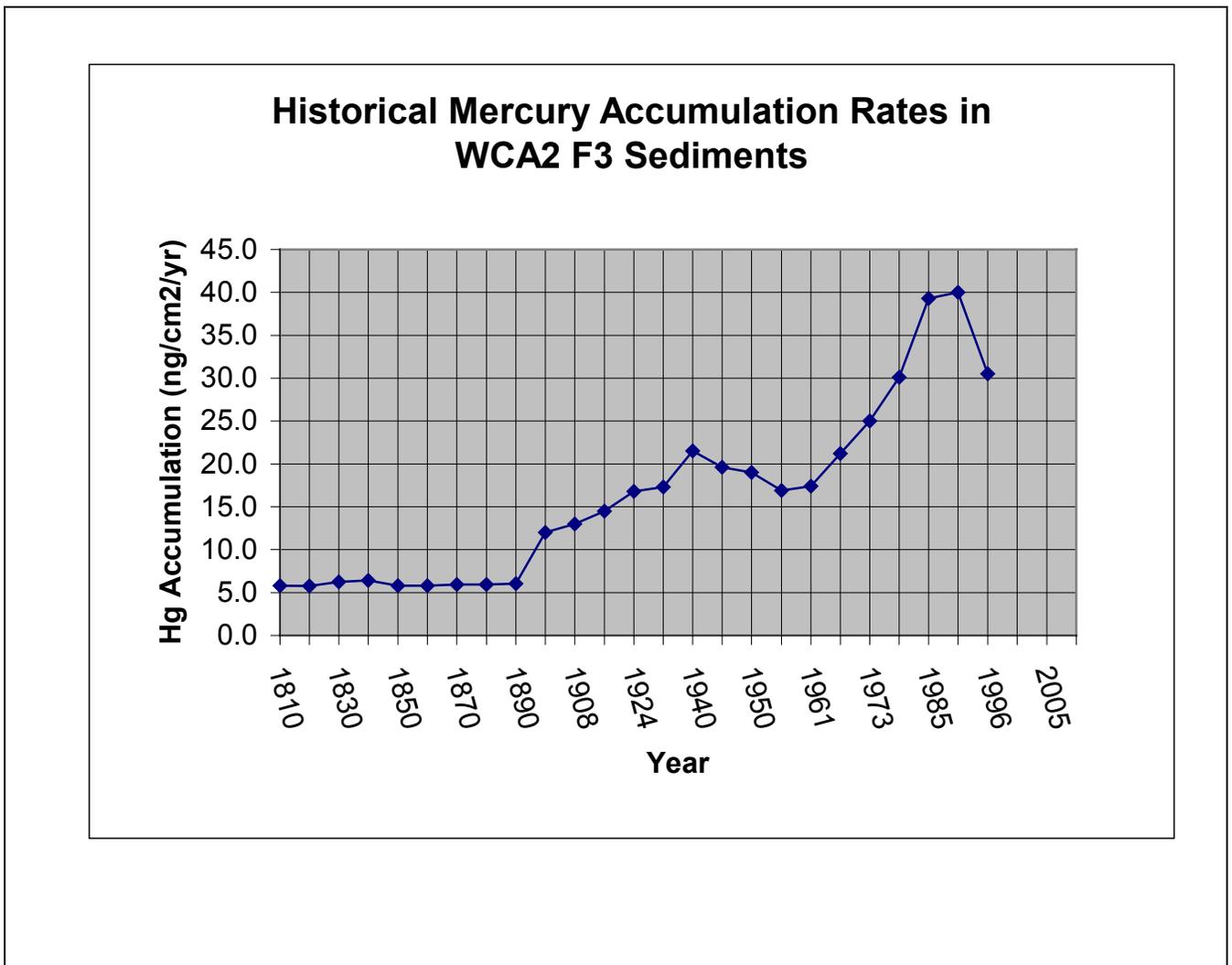


Figure 2B-7. Trends in mercury accumulation in Everglades soils at site F3 in WCA-2A. Mercury accumulation rates as determined by dated sediment cores are the best surrogate for trends in deposition in the absence of direct deposition measurements (Robbins, et al., unpublished data)

SUMMARY

In this section on the atmospheric cycling of mercury, the key features of this phenomenon have been described and the results of corrective measures taken by the FDEP and other agencies to minimize atmospheric releases of mercury have been provided. The preceding illustrations relate to the rewarding conclusions that control policy for atmospheric mercury has had the desired result in ameliorating the transport and deposition of mercury to South Florida and the Everglades and in reducing mercury body burdens in fish and wildlife.

THE AQUATIC CYCLE OF MERCURY

The pathways of mercury accumulation in fish and wildlife are complex. Though inorganic forms of mercury dominate its environmental cycle, a small percent of mercury can be transformed into methylmercury in waterbody sediments. Methylmercury is primarily produced by sulfate-reducing bacteria (SRB) naturally present in the sediment, where oxygen is absent and a sulfur compound (sulfate) is present. These bacteria take up inorganic mercury and convert it to methylmercury as an incidental byproduct of normal life processes. Other microorganisms living in the sediments or overlying water readily absorb methylmercury much faster than they excrete it. As other organisms feed on these microorganisms, methylmercury becomes progressively more concentrated at each higher level of the aquatic food web, a process known as bioaccumulation. This results in a buildup of methylmercury at each level of the aquatic food chain. In larger fish, levels of methylmercury can accumulate to as much as several million times higher than that of the surrounding water. In sufficient doses methylmercury is toxic to the brain, liver, kidney and immune system of animals and humans and can have adverse effects on egg and fetus development.

Many soil, water quality, and biotic factors directly or indirectly influence methylmercury production. For example, while sulfate is required for microbial methylmercury production, high sulfate levels tend to inhibit production. Drought and fire can increase the production of methylmercury by changing the proportions of sulfur forms in the soil, which can worsen the mercury problem locally over the short term. A better understanding of sulfur's role in mercury accumulation at sites with different levels of nutrient enrichment will permit agencies to evaluate the potential for minimizing the mercury problem through the management of water and its constituents. More detailed treatments of the general features of the environmental mercury cycle can be found in the 2000, 2001 and 2002 ECRs, and in **Appendices 2B-2** and **2B-5** of the 2003 ECR.

PHOSPHORUS AND MERCURY: THE ISSUE

Everglades Agricultural Area

The Everglades Agricultural Area (EAA) extends south from Lake Okeechobee (**Figure 1-1**) and comprises approximately 2,872 square kilometers (1,122 square miles) of highly productive agricultural land. Approximately 77 percent of the EAA consists of agricultural production. Nutrient-laden water from the EAA is recognized as a major contributor to Everglades nutrient enrichment.

Phosphorus from agricultural and urban development has been identified as the nutrient most responsible for changes in periphyton and plant communities within the Everglades Protection Area (EPA) (Koch and Reddy, 1992; McCormick and O'Dell, 1996; McCormick et al., 1998).

These changes include replacement with cattail of large areas once dominated by sawgrass and periphyton, a decline in wading bird populations, and species changes in periphyton and macroinvertebrate communities (Davis and Ogden, 1994; SFWMD, 1992a,b). Reducing phosphorus loading to the EPA from the EAA is central to the District's strategy for restoring and preserving the Everglades. Nutrient control is the primary focus of programs under the Everglades Forever Act (EFA).

Stormwater Treatment Areas

To address the phosphorus problem that results from EAA discharges, the initial management plan for the Everglades (SFWMD, 1992a) proposed the construction of three large water treatment wetlands encompassing approximately 14,500 hectares (ha), or about 35,000 acres (ac). These constructed wetlands, known as Stormwater Treatment Areas (STAs), were designed to serve as biological traps to reduce the phosphorus concentration in agricultural runoff entering the EPA and reduce the ecological effects of EAA discharges on the EPA. The interim EFA goal is for the STAs to achieve a total phosphorus (TP) level of 50 ppb. Advanced Treatment Technologies (ATTs) are being developed to further reduce phosphorus discharge from the EAA.

Sugar Cane Growers Cooperative

The Sugar Cane Growers Cooperative (SCGC) has expressed concern that reducing phosphorus concentrations in Everglades waters via STA treatment of the phosphorus-rich waters discharged from the EAA could exacerbate the Everglades mercury problem by reducing biodilution. Biodilution is an increase in biomass of plants due to increased phosphorus concentration, which may result in lower mercury concentration per unit of plant biomass and could possibly cause less bioaccumulation of mercury in fish. The SCGC's environmental consultants have opined that, "Controlling phosphorus from 30 ppb down to 10 ppb will cause mercury to increase in mosquitofish and wading birds, placing birds at increased risk." (PTI, Inc., 1994, 1995a, 1995b, 1997).

An increase in mercury in fish and birds would, of course, be a matter of concern. Two million acres of the Everglades are already impaired because of high mercury concentrations in sport fish. Health advisories have been issued encouraging the public to avoid or limit consumption of these fish.

The weight of evidence, however, does not support the contention that reducing phosphorus to near its natural concentrations in the Everglades to achieve ecological goals will greatly exacerbate the Everglades mercury problem.

Factors Influencing Mercury Concentrations in Everglades Fish

Methylmercury, the most toxic form of mercury, is produced from inorganic mercury, a more common, less toxic form, by sulfate reducing bacteria (SRB) in sediments in the absence of oxygen. Methylmercury is not only highly toxic, but it efficiently bioaccumulates, achieving higher concentrations in animals higher up the food chain.

Conditions favorable to a methylmercury problem include high inorganic mercury concentrations in rainfall, decaying plants, shallow waters and low flow, frequent drying and rewetting, and excess sulfate in runoff. The Everglades exhibits all these conditions.

Many soil, water quality, and biotic factors directly or indirectly influence methylmercury production. For example, while sulfate is required for microbial methylmercury production, high sulfide levels tend to inhibit production. Drought and fire can increase the production of methylmercury by changing the proportions of sulfur forms in the soil, which can worsen the mercury problem.

Empirical data and Everglades Mercury Cycling Model (E-MCM) results suggest that inorganic mercury supply to the Everglades, primarily from atmospheric deposition, is the ultimate cause of the mercury problem and the most important determinant of the methylmercury production rate and concentration in the Everglades. The proximate cause of the mercury problem is the production rate of the SRB that produce methylmercury. In the Everglades, SRB are mainly found in sediments, and SRB methylmercury production rate is closely linked to the concentration of methylmercury in fish.

Inorganic mercury supply to the Everglades is the ultimate cause of the mercury problem, a conclusion that is arrived at through common sense and by a hypothesis supported by research findings. Mesocosm dosing studies in the Everglades have shown that the addition of inorganic mercury into mesocosm surface waters resulted in methylmercury production within 1 to 2 days of dosing. Methylmercury produced from the added inorganic mercury accumulated rapidly in *Gambusia* (mosquitofish). The increase in methylmercury in surface sediments and in fish showed a linear response to the dose of inorganic mercury (Krabbenhoft and Gilmour, 2002).

The SCGC asserts that the process of reducing phosphorus in the EPA will exacerbate the mercury problem in the impacted area. The SCGC also argues that phosphorus control measures should be tempered. The view of the FDEP, the District, and most scientists is that the rate of conversion of inorganic mercury to methylmercury by SRB is much more a function of inorganic mercury, sulfate and organic carbon availability than phosphorus concentration. Phosphorus is not a direct determinant of methylmercury production rate. In Everglades sediments, where the bulk of mercury methylation occurs, environmentally realistic changes in phosphorus concentration will have little effect on SRB rate of production of methylmercury (**Figure 2B-8**).

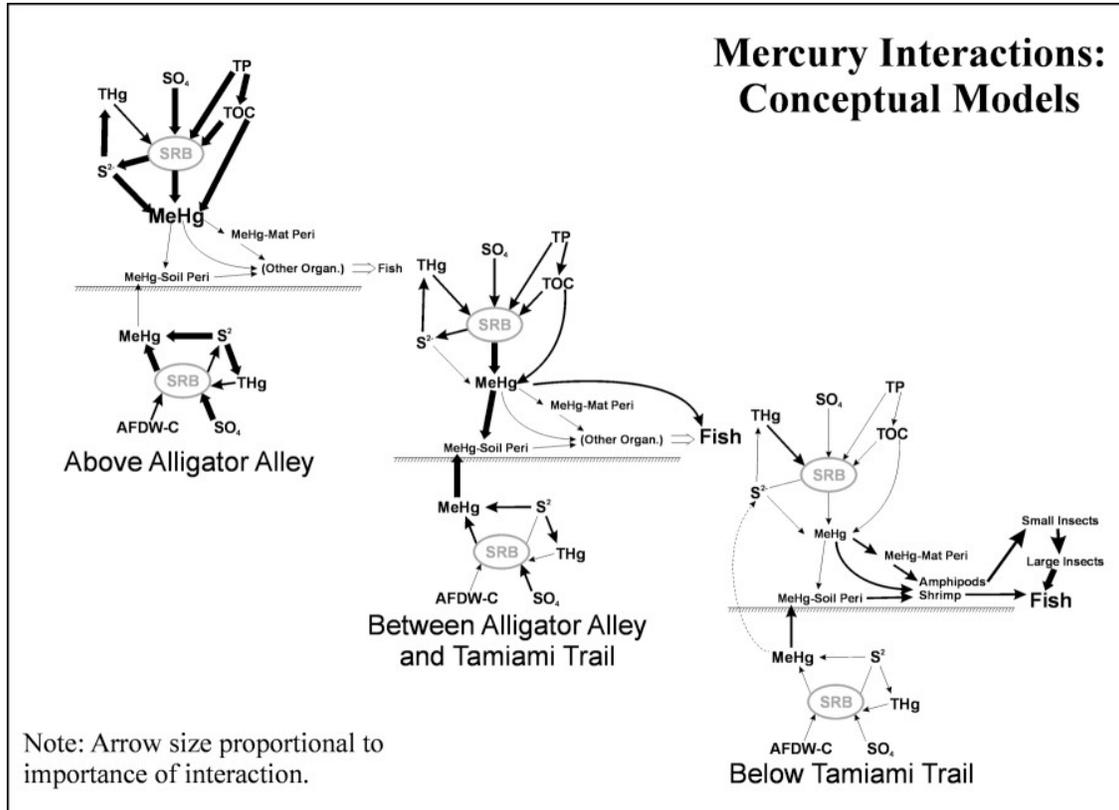


Figure 2B-8. The relative importance of water quality factors on methylmercury production and bioaccumulation in the northern, central and southern Everglades (Stober et al., 2001)

The Influence of Phosphorus on Methylmercury Production

Testing of the SCGC’s hypothesis, which asserts that higher phosphorus concentrations reduce methylmercury bioaccumulation in the Everglades, is complicated by the fact that phosphorus co-varies with other water quality parameters in the ecosystem. For example, there are significant north-to-south gradients from Lake Okeechobee to Florida Bay in total phosphorus (TP), total organic carbon and sulfate. Therefore, it is difficult to ascribe effects to phosphorus alone. Secondly, Stober et al. (2001) believe that high phosphorus concentrations exert a bottom-up control on ecosystem responses in the northern Everglades, while ecological interactions exert a top-down control on ecosystem responses in the low-nutrient southern area. The amount of organic carbon discharged from the EAA or produced by plants because of higher TP concentrations may overwhelm the system’s capacity to handle the volume of organic material. Dissolved oxygen (DO) concentrations are low or zero. This condition is not tolerated by many invertebrates and fish. The food chain, therefore, could be altered in this area so that mercury is not efficiently accumulated to the higher trophic levels. Thirdly, phosphorus affects many biogeochemical processes, including redox potential, detritus decomposition rate, and organic carbon supply, some of which may influence – positively or negatively – mercury methylation rate.

Several lines of evidence suggest that phosphorus is not a direct determinant of methylmercury concentration in Everglades fish.

Phosphorus Addition to Everglades Mesocosms

Krabbenhoft and Gilmour (2002) measured methylmercury concentrations and production rates in surface sediments across the Everglades from 1995 through 1998 as part of the ACME program. Methylmercury concentrations and production rates varied by more than 100 fold from eutrophic northern WCA-2A to the methylmercury maxima in central WCA-3A. To determine the factors responsible for this wide range of methylmercury production rates, Krabbenhoft and Gilmour employed mesocosms to study the effect of chemical amendments, including phosphate enrichment, on methylmercury production.

It was hypothesized that phosphate might directly influence net methylmercury production, either through effects on the growth of methylating and demethylating bacteria or by affecting the complexation, and therefore bioavailability, of mercury.

Phosphate-enrichment mesocosm studies were conducted at four sites in the Everglades over the last decade by Newman, McCormick and others at the District. Access to these experimental systems afforded ACME researchers the opportunity to examine phosphate's influence on methylmercury production separately from other factors, such as sulfate, that co-vary with phosphate across the Everglades. The four mesocosm sites exhibited *in situ* phosphate enrichment ranging from the moderately enriched site U3 in WCA-2A to more pristine sites in central WCA-3A, in central Arthur R. Marshall Loxahatchee National Wildlife Refuge (Refuge) and Taylor Slough in Everglades National Park (ENP). At the time of sampling, the mesocosms were at or near steady state with respect to plant responses to phosphate additions, providing an opportunity to examine effects of enhanced plant growth on net methylmercury production.

The phosphate addition mesocosm experiments revealed no direct effect of phosphate on net methylation (Gilmour et al., 2000). While phosphate enrichment significantly changed plant and periphyton communities in the mesocosms, phosphate enrichment changed methylmercury concentrations in surface sediments by less than a factor of three at any mesocosm site. Further, there was no trend across mesocosm sites in the direction of any methylmercury response to phosphate loading (Gilmour et al., 2001) (**Figure 2B-9**). To put these responses in context, methylmercury concentrations in surface sediments in mesocosms varied by less than a factor of three at any site in response to phosphate loading, compared with the more than 100-fold range in methylmercury concentrations and production rates across the Everglades from eutrophic northern WCA-2A to the methylmercury maxima in central WCA-3A. These *in situ* mesocosm studies confirm and extend smaller-scale studies that show little direct effect of phosphate on methylmercury production and accumulation in surface sediments. The studies also argue against biodilution as a mechanism that may reduce methylmercury bioaccumulation in the Everglades.

More likely, phosphate could indirectly affect net methylmercury production through enhanced plant growth, leading to higher organic carbon supply to sediment microorganisms and, possibly, changed redox conditions in sediments. The organic matter supply to sediments could affect microbial activity in sediments and would control sulfate-reduction and sulfide-production rates at locations where sulfate is not limiting. Further, dissolved organic carbon acts as a strong ligand both for mercury (Ravichandran, 1998; Ravichandran et al., 1999; Benoit et al., 2001) and for methylmercury (Hintelmann et al., 1995; Miller et al., 2001) and may inhibit the uptake of methylmercury into biota.

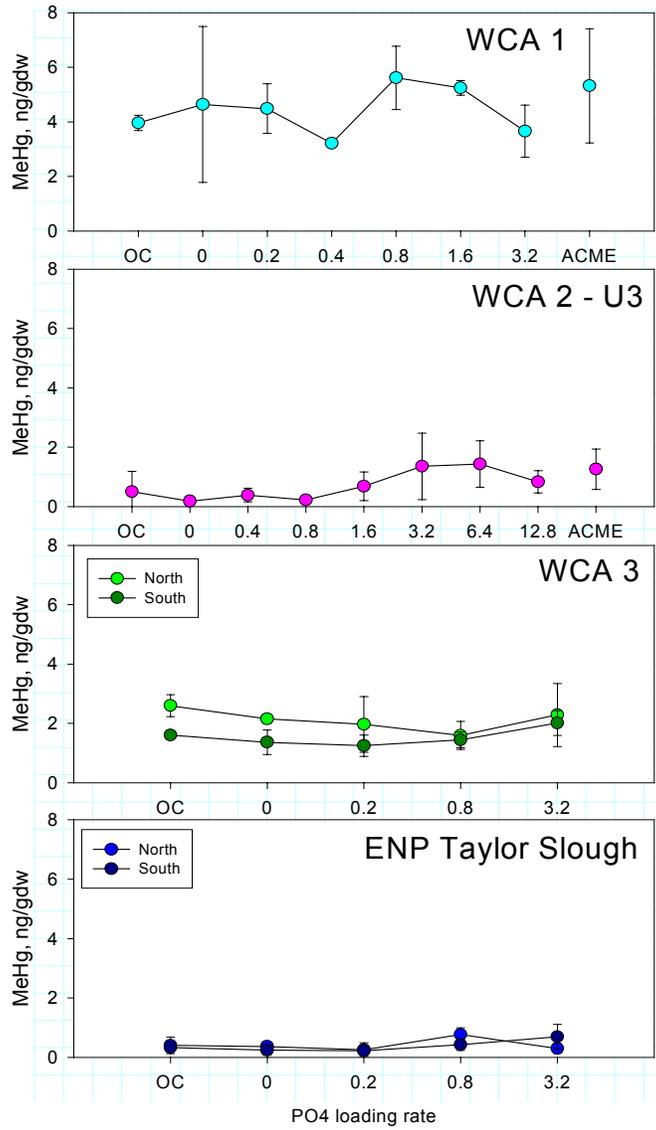


Figure 2B-9. Relative responses of mesocosms to phosphate addition. Mercury methylation rates vary by 100-fold across the Everglades, whereas phosphate additions result in a less than three-fold change

Nutrient effects on mercury cycling that are mediated through increased plant growth rates, resulting in higher organic carbon supply to sediment microorganisms, changed redox conditions in sediments, and altered sulfate reduction and sulfide production rates, should be examined over a longer term. However, it is difficult to argue that such systemic effects to the ecosystem would be advisable or acceptable as a potential but unproven means of reducing methylmercury bioaccumulation in the Everglades.

In contrast to the lack of methylmercury response to phosphate enrichment, sulfate stimulates methylmercury production when added to sediment cores taken from sites with lower sulfate concentrations (< 5 mg/L) (Bell, et al., 1999; Gilmour, et al., 2000; Gilmour, et al., 2001a, 2001b). In these studies, methylation rates increased linearly up to a point with sulfate addition, and then decreased with increasing pore water sulfide concentrations. The optimal sulfate concentration appears to vary somewhat among Everglades regions because of the way sediment chemistry affects the solubility of sulfide produced by SRB.

There is now evidence that sulfate and inorganic mercury inputs co-mediate bacterial production of methylmercury, thereby co-limiting bioaccumulation (Hrabik and Watras, 2002).

USEPA REMAP Data

The USEPA has been monitoring environmental conditions throughout the Everglades Protection Area (EPA) with its South Florida Ecosystem Assessment Project. This project uses a statistical, probability based sampling design to select sampling sites. All marsh locations within the EPA have an equal probability of being sampled. This design allows estimation, with known confidence and without bias, of the status of various indicators of ecological resources.

From 1993 to 1999, the USEPA Region 4 conducted the South Florida Ecosystem Assessment Project, commonly referred to as the Everglades REMAP (Regional Environmental Monitoring and Assessment Program) effort (Scheidt et al., 2000; Stober et al., 1996; 1998; 2001⁵). During 1995 through 1996, and also in 1999, about 750 marsh locations were sampled throughout the Everglades Protection Area (EPA), an effort that is unprecedented in terms of simultaneous spatial coverage and intensity.

Freshwater marsh stations were sampled within the EPA during 1995, 1996 and 1999, once during the dry season and once during the wet season in each of these years, for a total of six synoptic sampling events. Each synoptic event included about 125 stations for a total sampling effort of about 750 marsh sites. Sampling included water, soil, plants and fish. At each marsh location where water was present, mosquitofish (*Gambusia*) mercury was determined, along with surface water TP. (In addition, four transects along phosphorus gradients were sampled once during 1994).

A variety of statistical methods have been explored to determine whether there might be an association between surface water phosphorus and mosquitofish mercury. Analysis of this extensive data set (n = 545) failed to find a significant relationship between phosphorus in surface water and mercury in mosquitofish across the EPA using either linear or exponential regression. Eliminating all phosphorus data points above 30 ppb to examine the SCGC's hypothesis that controlling phosphorus from 30 ppb down to 10 ppb would cause mercury to increase in

⁵ (All reports are available online at: http://www.epa.gov/region4/sesd/sesdpub_completed.html)

mosquitofish again failed to find a relationship between mercury in mosquitofish and phosphorus ($p < 0.05$) (**Figure 2B-10**). Further analysis of the REMAP data set, including log normal transformations of data and running analyses on the northern Everglades and southern Everglades separately (north and south of the Tamiami Trail), failed to find a relationship between mercury in mosquitofish and phosphorus. This suggests there is no relationship between water phosphorus and fish mercury. Further, it is not possible to predict fish mercury concentration from water phosphorus. A more detailed analysis of such data appears in **Appendix 2B-5**.

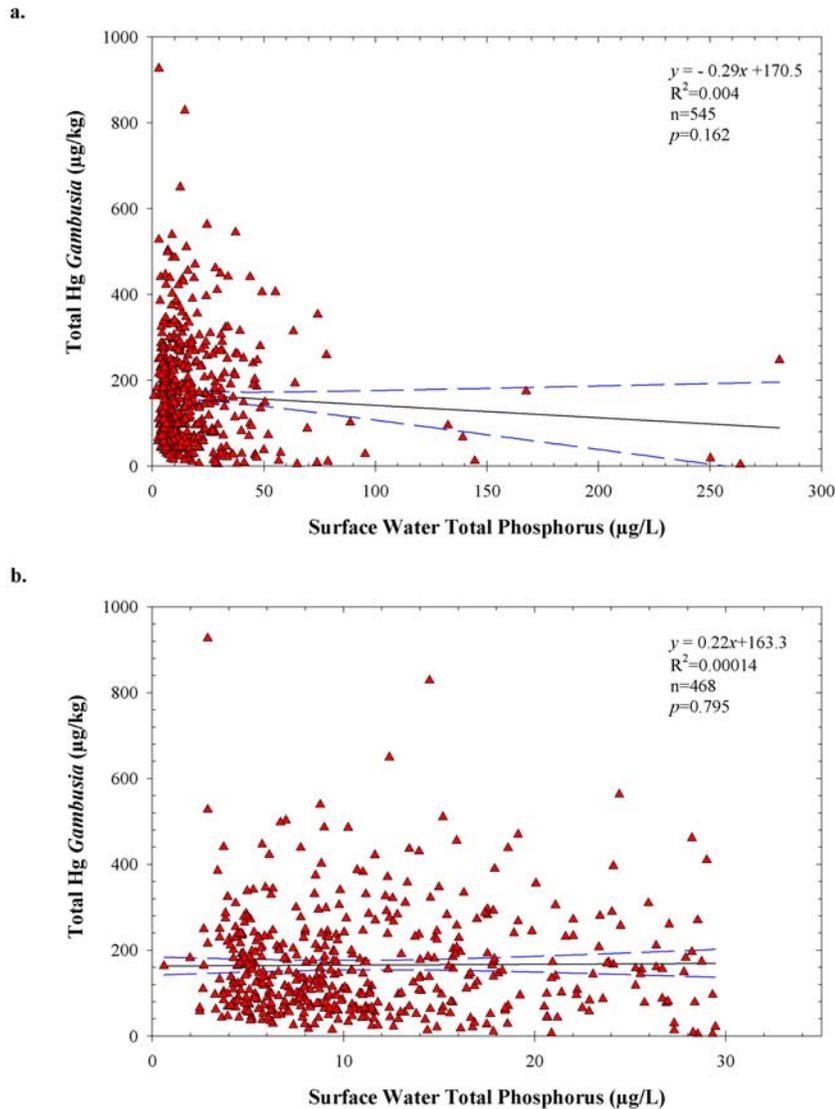


Figure 2B-10. Linear regression of THg in *Gambusia* (mosquitofish) versus TP in Everglades surface waters (data from USEPA REMAP). Figure 2B-11a, all data. Figure 2B-11b, TP data > 30 ppb excluded

METHYLMERCURY AND PHOSPHORUS RELATIONSHIPS IN STA-2

STAs can effectively remove phosphorus from EAA inflow waters. STA-1W, STA-5 and STA-6 have average inflow TP concentrations of about 125 ppb, 210 ppb, and 77 ppb, respectively, while outflow concentrations average about 35 ppb, 130 ppb, and 30 ppb, respectively. **Figure 2B-11** indicates STAs can be managed to minimize methylmercury production, as some STAs are removing phosphorus while essentially achieving no net production of methylmercury.

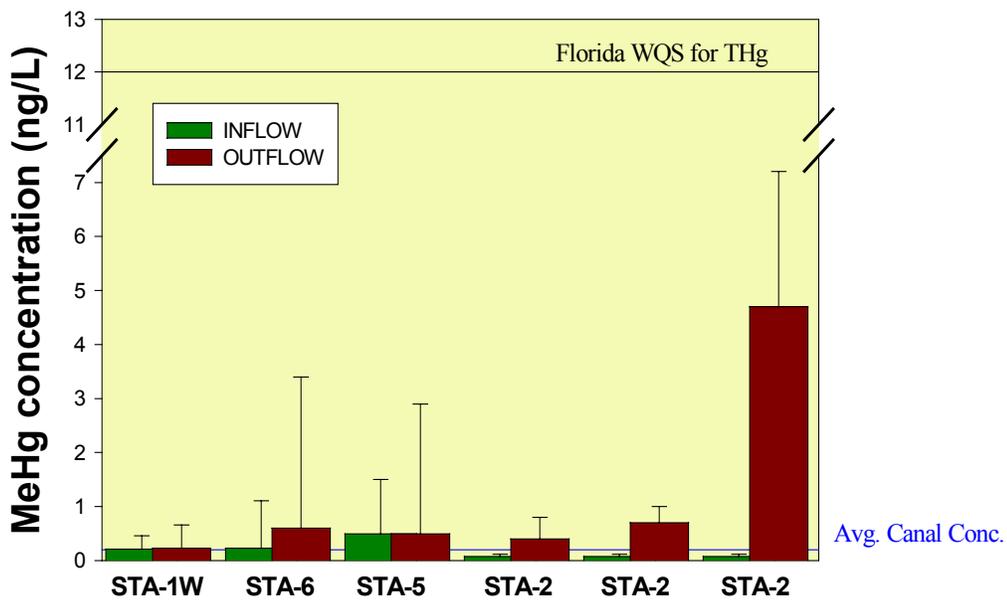


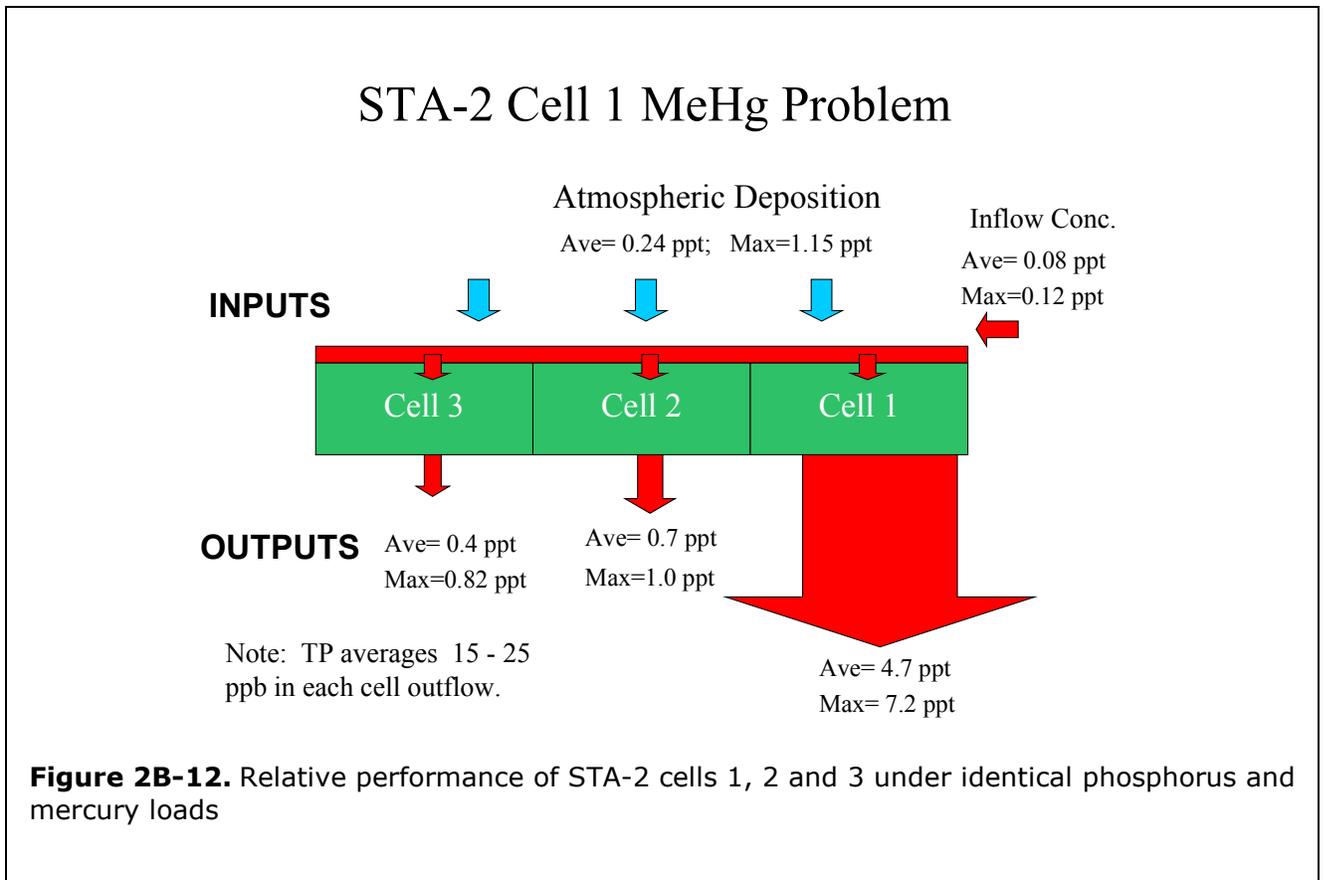
Figure 2B-11. Methylmercury inflows and outflows of the STAs

From fall 2000 to the present, STA-2 has experienced several anomalous mercury events in which methylmercury concentrations in the interior or outflow waters were in the range of 10 to 100 times the inflow concentration. These events are confined to cell 1 and have occurred about 90 days after summer reflooding following extended periods of drawdown and dryout in the dry, winter months. Moreover, these anomalies are not confined to the water column. The differences in the average total mercury concentration in mosquitofish between cell 1 and cells 2 and 3 are equally striking. While cell-1 mosquitofish averaged 0.18 ppm (wet weight) in the fall collection

in 2001, cell 2 and 3 fish averaged 0.069 ppm (wet weight) and 0.010 ppm (wet weight), respectively. These differences are also propagating up the food chain to sunfish and bass, but the annual drawdown of cell 1 has prevented the development of a resident population of the large-bodied top-predator fish. Consequently, between-cell comparisons are not as clear.

The concentration of TP at the S-6 pump station, which pumps water into the STA-2 supply canal, averages about 70 ppb. The supply canal provides virtually the same water quality to each cell. While each cell is operated independently, the average outflow concentrations of TP are from 15 to 25 ppb, with virtually indistinguishable differences between cells. This indicates that the interior concentrations of water column TP are also virtually indistinguishable, yet the methylmercury behavior of cell 1 is distinctly different from that of cells 2 and 3.

Clearly, TP is not causing or contributing to a mercury problem in STA-2 cell 1 because cells 2 and 3, which have no mercury problem, have exactly the same TP concentrations in water and soil as cell 1 (**Figure 2B-12**). It must follow, then, that something other than TP is driving these differences in mercury behavior. One of the differences is that cell 1 dries out and rewets more frequently than do cells 2 and 3. The effect of drying and rewetting on the mercury cycle in the Everglades is addressed in **Appendix 2B-1**. Another difference lies in the cell 1 soil chemistry. The influence of soil chemistry before and after dryout and rewetting on the mercury cycle is also discussed in **Appendix 2B-1**.



While the District, the FDEP and the USEPA have concluded that the STA-2 cell 1 mercury problem is not related to TP, all three agencies are taking the anomalous mercury behavior of STA-2 cell 1 seriously. A series of studies is now under way to probe the differences among cells 1, 2 and 3 that are causing or contributing to the cell 1 anomalous mercury behavior. One laboratory study is looking at the effect of drying and rewetting on sulfur and mercury chemistries in surface water, soil pore water, and soil solids over time using soil cores collected from cell 1. Another study is planned to evaluate the effect of sulfate loading from inflow canal water on sulfur and mercury chemistry in the inundated soils of cell 1 using a set of replicate field enclosures (mesocosms). A third study will increase the number of sites and parameters to be monitored and the frequency of monitoring to ensure that the influences of inflow, rain quantity, quality, and water depth, drying, and rewetting are tracked more closely than would otherwise occur with routine monitoring. Finally, the revised Everglades Mercury Cycling Model (II) will be adapted to STA-2 and its downstream environment to evaluate the effect of various operational scenarios on excess MeHg production and bioaccumulation within and downstream. Over the next three years the District's and the FDEP's combined investment to understand and mitigate the STA-2 cell 1 mercury problem is expected to be approximately \$1 million.

BIODILUTION

The proposed TP water quality criterion of 10 ppb is intended to restore and protect the natural Everglades. The Sugar Cane Growers Cooperative of Florida (SCGC) has raised concerns that reducing phosphorus concentrations in Everglades waters to this proposed numerical water quality criterion could worsen the Everglades mercury problem by reducing biodilution. Biodilution results from a sustained increase in plant production due to a sustained increase in the limiting factor to plant production (e.g., light and nutrients).

All other factors being equal, biodilution causes a decrease in methylmercury concentrations per unit of plant biomass, a decrease in the average concentrations of methylmercury in organisms that feed on plants, a decrease in the concentrations of methylmercury in the predators of the organisms that feed on plant grazers, and so on up the aquatic food chain.

In updating its concerns regarding the alleged inverse relationship between phosphorus concentration in water and methylmercury concentration in upper trophic-level animals, the SCGC has in particular referred to a recent publication by Pickhardt et al. (2002).

Pickhardt and co-workers tested the hypothesis that at equal, initial concentrations of methylmercury dissolved in water in "tanks" or aquaria, an increase in the cell density of a population of one-celled plants (algae) would result in a decrease in the methylmercury concentration per algal cell. In this case, the increase in algal densities in aquaria was caused by an increase in the phosphorus concentration. The rationale for this hypothesis was that the concentration of methylmercury per algal cell would be lower in denser algae blooms because the same amount of methylmercury would be distributed among a greater number of algal cells.

Pickhardt and co-workers, after allowing the methylmercury time to distribute among the algal cells in each aquarium, added water fleas (*Daphnia* sp.) to the aquaria. Water fleas, a member of a category of organisms referred to as zooplankton, feed on algal cells. This is the simplest food chain that can be constructed for such studies. By extension to the hypothesis that methylmercury per algal cell would decline with increasing densities of algae, it was further hypothesized that water fleas feeding on algae cells would assimilate less methylmercury from aquaria with higher densities of algae than from aquaria with lower densities of algae, all other things being equal.

The results of Pickhardt et al. indicate a biodilution effect over the range of phosphorus concentrations from 7.4 µg/L to 44.6 µg/L phosphorus. It is not surprising that there was a biodilution effect in this simple experimental system over a six-fold range in phosphorus concentration. For the Everglades, however, phosphorus concentration must not cause an imbalance of flora and fauna. Consider that the FDEP's proposed numeric water quality criterion for phosphorus is 10 ppb to avoid imbalance, and the criterion proposed by the Sugar Cane Growers Cooperative is 15.6 ppb phosphorus to avoid imbalance. Data from Pickhardt et al. demonstrate that the biodilution effect over this 10-ppb to 15.6-ppb phosphorus range is small. The Pickhardt et al. regression line indicates a 13 percent reduction in methylmercury bioaccumulation in algae, and similar small declines in bioaccumulation in water fleas over the 10-ppb to 15.6-ppb phosphorus concentration range.

It would appear that even under these ideal conditions (algae and an algal grazer contained in aquaria), the observed biodilution effect over the phosphorus concentration range being considered for the Everglades phosphorus criterion was small. In addition, there are problems associated with extrapolating this trend to real aquatic ecosystems in general or to the Everglades in particular. The Pickhardt et al. experimental design was intended to simulate conditions that occur primarily in deep lakes. No sediment was added to aquaria, there was no significant biological means for methylmercury to be produced from inorganic mercury in the aquaria, there were no periphyton or floating or rooted aquatic macrophytes in the aquaria, and there was no opportunity for the simple phytoplankton and zooplankton food web in the aquaria to be altered as to animal species or number of trophic levels present as a consequence of increased phosphorus addition.

As such, the Pickhardt et al. experimental design may have simulated some of the conditions in deepwater lakes, but it could not have simulated the conditions in the shallow wetlands of the Everglades, nor was it designed to do so. There are several important differences between deepwater lakes and the Everglades that must be acknowledged. First, the Everglades does not generally respond to additional phosphorus loading by producing higher densities of free-floating algal cells (phytoplankton) or the strings of algae that form dense mats (periphyton). In fact, in the most eutrophic portions of the Everglades, the cattail canopy is so dense it shades out the periphyton mats that would otherwise flourish under the high-phosphorus concentrations. Second, processes other than biodilution affect the degree of bioaccumulation of methylmercury to upper trophic levels in the Everglades; some of these processes are influenced by phosphorus, while others are not.

One of the other processes affecting the degree of bioaccumulation of methylmercury to upper trophic levels is methylmercury production rate. In the Pickhardt et al. aquaria experiment, methylmercury supply was, by design, not affected by phosphorus concentration. In the Everglades, however, phosphorus additions could affect organic carbon production and sulfur dynamics and therefore might either increase or decrease the methylmercury production rate by SRB through indirect effects. In addition, methylmercury production has been measured in the thick periphyton mats at the most phosphorus-impacted research site in WCA-2A, but not at the unimpacted research site. This effect on overall methylmercury production is muted, however, because the periphyton mats occur only sparsely at the former site due to cattail shading.

Water chemistry is a second factor affecting methylmercury bioaccumulation. For example, dissolved organic carbon (DOC) is known to compete with algae for methylmercury and weaken the biodilution effect. DOC was not monitored or controlled in this study; however, it is unlikely that DOC concentrations approached the high levels typical of the Everglades.

A third factor affecting methylmercury bioaccumulation is food web structure. The degree of bioaccumulation to upper trophic levels is clearly affected by changes in food webs, and phosphorus is a factor that can dramatically alter food webs. There was no opportunity for the aquaria food web in the Pickhardt et al. experiment to be altered as to species or number of trophic levels present in response to increased phosphorus addition. However, it is obvious that the Everglades food web is dramatically altered by increases in phosphorus loading.

Based on the above, the biodilution results of Pickhardt et al. cannot be extrapolated to the Everglades. In any case, it is clear from several lines of evidence previously presented that phosphorus is a poor predictor of methylmercury production and bioaccumulation in the Everglades. The biodilution issue is discussed in greater detail in **Appendix 2B-5**.

THE INFLUENCE OF PHOSPHORUS ON METHYLMERCURY BIOACCUMULATION

The Likely Effect of the Everglades Construction Project on the Everglades Mercury Problem

In 1993 the Sugar Cane Growers Cooperative (SCGC) first voiced its concern that reducing water column TP might exacerbate the mercury problem in receiving waters by reducing the biodilution associated with eutrophication. Support for this concern came from experience elsewhere with lakes and from a limited data set that showed an increase in methylmercury bioaccumulation in mosquitofish within WCA-2A that correlated inversely with the concentration of TP in the water column.

Based on the latter, PTI Environmental Services (under contract to the SCGC) developed a one-variable model to predict mercury in mosquitofish based on TP concentrations in the water column. Subsequently, PTI (and later, Exponent) prepared a series of reports that used three models in combination to generate a prediction of the post-STA risks of methylmercury toxicity to wading birds feeding exclusively in the restored area in WCA-2A downstream of the S-10 structures (PTI, 1995a,b; PTI, 1997; Exponent, 1998). Results of the risk assessments will be discussed in the following section. This section summarizes a previously published evaluation of Exponent's empirical model, which it used to predict mercury bioaccumulation in mosquitofish in response to phosphorus reduction in the most impacted Everglades areas. This section also references the results of a multivariable mechanistic model that supports the District's conclusion that post-restoration mercury risks will be acceptable.

In Appendix 7-4 of the 1999 *Everglades Interim Report*, the District carried out a detailed evaluation of PTI's empirical model to predict the effect of phosphorus reduction on mercury bioaccumulation in the Everglades food chain. For this exercise, the District pursued two lines of reasoning. The first involved evaluating the validity of Exponent's empirical model by comparing its predictions to those generated by other empirical models of equal or greater statistical reliability. The second involved evaluating the validity of Exponent's empirical model by comparing its predictions to those generated by the USEPA's mechanistic mathematical model, which was initialized and calibrated with five years of monitoring and research data collected along the nutrient gradient in WCA-2A.

Based on the first line of reasoning, the District derived several other one- and two-variable empirical models that were better predictors of methylmercury in mosquitofish than the model using only water column TP. Nevertheless, the District concluded that neither Exponent's

one-variable empirical model based on TP, nor the District's two-variable model based on DOC and calcium, should be used to predict post-STA methylmercury concentrations in fish for ecological risk assessment because neither model could be demonstrated to have predictive value beyond the data sets used in their derivation. Moreover, since the STAs would reduce TP and total mercury, but not DOC and calcium, their ratios would change in ways that had not yet occurred and therefore could not yet be captured by a statistical model. The District rejected the predictive value of any empirical model for this application.

To pursue the second line of reasoning the District asked the USEPA's Office of Research and Development (ORD) to evaluate the post-STA effect of a reduction in biodilution when water column TP declined from an average 50 ppb to an average 10 ppb. The potential benefit of a post-STA 50-percent reduction in the total mercury load was also evaluated for each phosphorus scenario. The model results indicated that the expected downstream increase in mosquitofish MeHg that could be attributed solely to a reduction in biodilution was about 50 percent, not the 660 percent predicted by Exponent's empirical model. When the benefit of a potential 50 percent reduction in the EAA mercury load was added, the increase declined to about 40 percent.

Though not definitive, the results of this mechanistic modeling analysis strongly indicated that Exponent's empirical model was likely substantially overestimating the post-STA increase in mosquitofish MeHg concentrations attributable to a reduction in biodilution. Nothing that has been published or learned since then has altered the FDEP's position on this issue. To the contrary the results of the more recent application of the Everglades Mercury Cycling Model (E-MCM) Version II reinforce the FDEP's earlier conclusions.

In addition, the mechanistic modeling results led to the conclusion that there was likely to be a substantial margin of safety in the District's worst-case estimate of the methylmercury risks to wading birds feeding exclusively in the restored areas of the Everglades when the surface water phosphorus concentration is reduced from present-day levels to the proposed 10 ppb Water Quality Standard (WQS). Results of the District's worst-case ecological risk assessment are summarized in a subsequent section of this chapter.

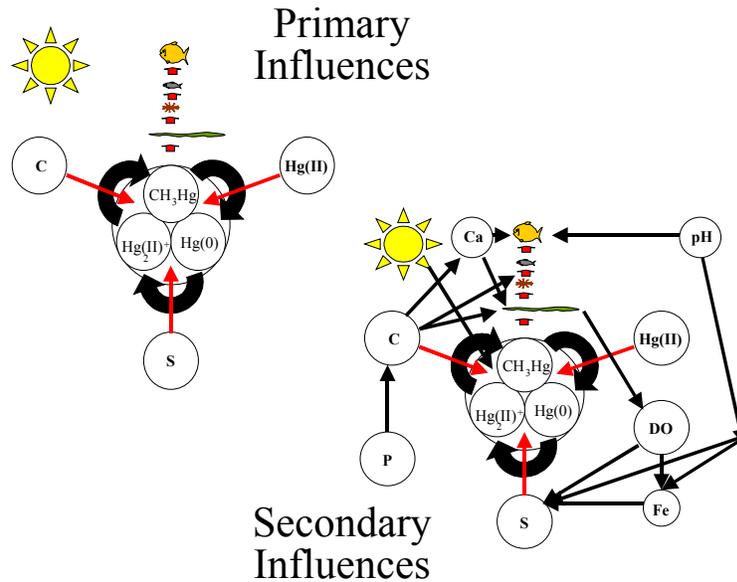


Figure 2B-13. Summary of primary and secondary influences on methylmercury production and bioaccumulation

Key Findings Regarding the Aquatic Cycle of Mercury in the Everglades

Summarized below are key findings of the United States Geological Survey (USGS) Aquatic Cycling of Mercury (ACME) Study Phase 1 (1995 through 1999), which focused on methods development and field monitoring, and the preliminary findings of Phase 2 (2001 through 2003), which focuses on mesocosm dosing studies to measure the effect of the inorganic mercury, sulfur, carbon, and phosphorus loading rates on methylmercury production and bioaccumulation in the Everglades environment rather than in a laboratory:

- SRB convert inorganic mercury from the atmosphere or from surface water inflows to methylmercury over a period of hours to days.
- MeHg production is the primary determinant of MeHg bioaccumulation in the Everglades.
- Sulfur chemistry is the primary determinant of MeHg production in the Everglades, but carbon chemistry also appears to play an important role.
- Drying and rewetting have a primary influence on sulfur chemistry.

- Phosphorus chemistry has a secondary influence on sulfur chemistry via the C, O, and Fe/Mn cycles.
- Food web structure has a primary influence on MeHg bioaccumulation.
- Everglades water chemistry and hydrology have a complex influence on food web structure.
- Biodilution, a consequence of excess primary production, is a secondary determinant of MeHg bioaccumulation.
- Biodilution also has a secondary influence on MeHg production by increasing peat formation rate and diluting soil inorganic mercury.
- Phosphorus controls primary production in most of the Everglades, except where the cattail canopy is so dense that light, not phosphorus, limits primary production. Ironically, this occurs in the most eutrophic areas of the Everglades, so the link between phosphorus and biodilution is broken precisely where it would be expected to be most evident.

SUMMARY

- Atmospheric deposition of inorganic mercury is the ultimate cause of the Everglades mercury problem, and the methylmercury production rate of SRB is the proximate determinant of the concentration of methylmercury in fish in the Everglades.
- Sulfur follows inorganic mercury deposition rate in order of importance regarding factors controlling methylmercury concentration in Everglades fish. Sulfur is a primary controller of the methylmercury production rate by SRB.
- Phosphorus plays a secondary role by influencing the carbon, oxygen, and sulfur cycles that affect the SRB methylmercury production rate.
- Phosphorus has no discernible direct effect on methylmercury production. Data from the USEPA REMAP program, from STA-2 monitoring and from phosphate addition to mesocosm experiments indicate that water column phosphorus concentration does not significantly correlate with methylmercury production.
- The Sugar Cane Growers Cooperative contends that reducing Everglades phosphorus concentrations to near their natural historical levels via treatment of phosphorus-rich discharge waters from the EAA by means of STAs and ATTs could exacerbate the Everglades mercury problem by reducing biodilution.
- However, the relationship between Everglades phosphorus concentration and methylmercury production rate is indirect and weak. While increased phosphorus concentrations could theoretically increase biodilution and reduce mercury concentrations in fish, phosphorus also alters sulfur, organic carbon and oxygen concentrations and speciation, which could act either to increase or decrease the methylmercury production rate. Additionally, increased phosphorus concentration alters Everglades plant communities and the entire food web, which may alter the level of bioaccumulation in fish.
- Though by no means definitive, the results of mechanistic modeling analysis strongly indicated that Exponent's (the environmental consultant to the SCGC) empirical model was likely substantially overestimating the post-STA increase in

mosquitofish methylmercury concentrations attributable to a reduction in biodilution.

- Phosphorus is neither a sensible nor an effective means of managing the Everglades methylmercury problem because phosphorus acts systemically, profoundly altering the Everglades ecosystem. The side effects of phosphorus far outweigh its potential, though unproven benefit as a “prescription” for reducing methylmercury.
- In contrast, source reduction of mercury from South Florida incineration sources has resulted in declines in mercury concentrations in Everglades biota in recent years and has resulted in positive effects for the ecosystem. Source reduction control, in contrast to allowing high phosphorus concentration inputs to the Everglades, does not add chemicals to the ecosystem but instead reduces their input. The most effective and reliable solution to the Everglades mercury problem is air source reduction, not manipulation of water quality. Mercury source reduction is by far the preferred means of managing the Everglades mercury problem.

ECOLOGICAL RISK TO EVERGLADES WILDLIFE

Assessment of Methylmercury Risks to Everglades Wildlife

The concern for potential mercury risks to wildlife began shortly after the first findings of high levels of mercury in largemouth bass in 1989 and the subsequent issuing of health advisories to fishermen. In late spring 1989, three Florida panthers died in eastern Everglades National Park. Florida Fish and Wildlife Conservation Commission (FWC) and U.S. Fish and Wildlife Service (USFWS) scientists performed a necropsy on each animal. Tissue samples were analyzed and were found to contain very high mercury concentrations. These findings stimulated concern that many forms of Everglades wildlife might be at risk for mercury toxicity.

In early planning sessions the SFMSP identified Everglades wading birds as being potentially at greatest risk for mercury toxicity. Not only had wading bird populations declined sharply in the Everglades during preceding decades, but studies of contaminant problems elsewhere often found birds to be at greatest risk of harm. Consequently the SFMSP sponsored a series of field and laboratory studies and experiments to define the potential risk of mercury to wading birds.

As discussed earlier, PTI Environmental Services (and later, Exponent, both of which were under contract to the Sugar Cane Growers Cooperative of Florida) prepared a series of reports predicting very high post-STA methylmercury risks to wading birds based on model predictions (PTI, 1994, 1995a,b; 1997; Exponent, 1998). If Exponent’s predictions were accurate, Everglades wood storks were ingesting almost nine times the amount of methylmercury considered safe for birds. The hazard estimate, or hazard quotient (HQ) for great blue herons was even higher at 14 times the safe daily dose.

Though never peer reviewed, these reports were submitted for consideration in a number of policy and administrative venues, including a formal challenge to the Everglades Nutrient Removal Project permit. Following a presentation by PTI to the January 1997 meeting of the Environmental Regulation Commission (ERC), it was the ERC’s sense that the Exponent risk assessment should be peer reviewed prior to use in the Everglades restoration decision-making process.

To address concerns raised by PTI, and later Exponent, in 1998 the District carried out a deterministic ecological risk assessment of the potential impact the Everglades Construction Project (ECP) might have on the Everglades mercury problem (i.e., based on point estimates of average and 95th percentile concentrations). That assessment tested the hypothesis that risk to wildlife from methylmercury would increase because phosphorus removal would reduce the extent of eutrophication in the Everglades marsh downstream of the STAs. To test this hypothesis the District assessed methylmercury exposure to great egrets, great blue herons and wood storks at two sites. The first site, a nutrient-unimpacted reference site in WCA-2A in the northern Everglades basin, was believed to be representative of future “post-ECP” oligotrophic conditions in the restored areas (TP concentrations average about 7.3 ppb at this site; SFWMD, unpublished). The second site, a methylmercury “hot spot” in the center of WCA-3A, was chosen as a positive control where some adverse effects were known or suspected. In both cases it was assumed that the birds fed only at the study site. This exposure assumption was adopted to ensure that all exposure estimates were biased high. Following USEPA guidelines, the District quantified the methylmercury risks to wading birds using measured rather than modeled concentrations of mercury in the fish most likely to be eaten by wading birds. The District’s post-ECP ecological risk assessment produced hazard quotients of about one-tenth those predicted by Exponent models (1998) for the wood storks, great egrets and great blue herons.

After evaluating multiple lines of evidence, the District concluded that methylmercury risks to wading birds feeding at the WCA-2A reference site were not unacceptable. The District further concluded that this constituted reasonable assurance that the STAs would not cause or contribute to an increase in the biological imbalance in the already impacted areas in the northern Everglades, and that the known benefits of phosphorus reduction outweighed the potential detriments of methylmercury increase should an increase occur. The latter highlights an important aspect of risk assessments, that is, that risk assessment should recognize all potential stressors (e.g., chemical, habitat loss or degradation) to provide a basis for comparing, ranking and prioritizing risks. In this way, comparative analyses can help focus more significant ecologically based risk management decisions on those stressors that pose the greatest risk to fish and wildlife. The results of this assessment were published in the *Everglades Interim Report* (Rumbold et al., 1999).

The following year, based on comments received from Exponent, the peer review panel for the District’s *2000 Everglades Consolidated Report* (2000 ECR) requested that District staff re-evaluate methylmercury risk and consider newly published information on toxic effects to great egrets. The panel felt that data from a number of sources indicated that exposure to young birds could put them at risk, and further suggested that a probabilistic risk assessment for mercury risks be performed.

To address the panel’s concerns, the FDEP and the District first organized a peer-review workshop along with the University of Florida to address toxic effects assessments for methylmercury in fish-eating birds (October 6 through 7, 1999, River Ranch, Florida). The expert panel consisted of Drs. G. Suter (USEPA), J. Ogden (SFWMD), M. Meyer (Wisconsin Department of Natural Resources), M. Wolfe (UC Davis), and D. DeAngelis (USGS). Workshop attendees reached two points of consensus during that workshop:

1. Until such time that a more sensitive or more appropriate toxic effects measure is published for fish-eating wildlife, studies done on the mallard, which was the basis of the District’s risk assessment, should be used in the derivation of acceptable daily dose.
2. Risk assessments of methylmercury should not continue to simply rely on the mallard studies; instead, other species, receptors and endpoints should be studied with greater rigor (Suter,

1999). Accordingly, when the District reiterated its risk assessment, this time using a probabilistic approach, it included an assessment of risk to nestlings in addition to assessing risk to the embryo.

The results of the District's probabilistic ecological risk assessment were published in the 2000 ECR (Rumbold, 2000) and confirmed the results of the earlier deterministic risk assessment, concluding the following:

- Although certainly not trivial, the baseline methylmercury risks to wading birds feeding exclusively at WCA-2A-U3 (i.e., reference site for post-ECP conditions) are considered acceptable.
- Therefore, restoring nutrient-impacted areas in the northern WCAs to the conditions that now exist at WCA-2A-U3 is unlikely to be a threat to wading birds feeding in those areas, i.e., the methylmercury risks would be acceptable.
- Alternatively, methylmercury risks to wading birds feeding exclusively in the WCA-3A methylmercury "hot spot" (i.e., WCA-3A-15) are of potential concern and warrant further study. Some studies are already underway. However, this area is presently minimally affected by EAA discharges, a situation that is not expected to change as a result of the ECP.

Exponent also reiterated its risk assessment, this time using a probabilistic approach and, more importantly, using mercury levels measured in fish rather than those predicted from models. Exponent's risk assessment was later published in the 2001 ECR, along with comments on the District's assessment (Exponent 2000, Appendix 1-2d, 2001 ECR). It should be pointed out that Exponent's new risk assessment reported hazard quotients (HQs) very similar to those the District reported (i.e., one-tenth of those predicted by PTI, 1995a,b), and that the remaining differences between the two risk assessments appeared to center more on interpretation and communication of risk (for the District's responses to Exponent's comments, see Appendix 1 through 3g in the 2001 ECR). Both risk assessments were then provided to a special mercury peer review panel for the 2001 ECR. While recognizing that additional studies must be performed, this peer review panel stated in its report that, "The SFWMD is to be commended for its use of probabilistic risk assessment for the wading birds. This is the current, cutting-edge risk methodology, and the assumptions and parameters used in these assessments are sound and reasonable." (Appendix 1-1a: Public and Peer Review Panel Comments, 2001 ECR).

Nevertheless, as stated above, the special mercury peer review panel for the 2001 ECR also recommended that the FDEP and the District continue researching methylmercury risk to Everglades wildlife, including further studies on effects to wading birds, as well as risk assessments for other wildlife species, including alligators and bats. While such efforts have been hampered by resource constraints, both the FDEP and the District have continued their studies to reduce risk uncertainties (i.e., through funding by the FDEP and in-kind service by the District). For example, in addition to routine, permit-mandated monitoring of mercury in fish tissues and egret nestling feathers, the District continues to collect egret eggs to better assess wading bird exposure and risk (for details, see Rumbold et al. 2001 and Appendix 2B-3 of the 2003 ECR). Further, the District has been assisting the USGS in a study to establish a critical egg concentration for various wading birds species in Florida. To assist the USGS, the District collected 168 eggs of five species (47 great egret eggs, 29 anhinga eggs, 58 white ibis eggs, 21 tricolor heron eggs and 13 snowy egret eggs) in 2001 and shipped them live to USGS-Patuxent (Laurel, Maryland), where they were incubated after being injected with MeHg. Preliminary results from that study suggest that the embryos of some species of fish-eating birds may be more sensitive to MeHg than the eggs of mallards, and that estimates of harmful levels of mercury in

eggs, which have been based on reproductive trials with mallards in the lab, may have to be re-evaluated (Heinz et al., 2001). Furthermore, District staff recently reported the results of a study examining mercury bioaccumulation in alligators as part of a larger, multi-agency study investigating the status of alligator populations in the Everglades (Rumbold et al., in press). Through comparisons with previously published studies, that report drew the following conclusions: (1) mercury levels have declined in some Everglades alligators since 1994, and (2) mercury levels do not appear to be a threat to the alligators.

Though no new, formal ecological risk assessment was carried out during the reporting year, field monitoring and research continue to refine knowledge of mercury bioavailability and food web biomagnification. Such information is key to developing an improved understanding of the food web relationships among mercury in prey and predator, and to supporting further refinements in the estimation of ecological risk.

The concern for potential mercury risks to wildlife began shortly after the first findings of high levels of mercury in largemouth bass in 1989 and the issuance of health advisories to fishermen. In late spring 1989, three Florida panthers died in eastern Everglades National Park. Florida Fish and Wildlife Conservation Commission (FWC) and USFWS scientists performed a necropsy on each of the animals. Tissue samples from each were analyzed and were found to contain very high concentrations of mercury. These findings stimulated concern that many forms of Everglades wildlife might be at risk for mercury toxicity.

In early planning sessions the SFMSP identified Everglades wading birds as one resource potentially at greatest risk for mercury toxicity. Not only had wading bird populations declined sharply in the Everglades during preceding decades, but also studies of contaminant problems elsewhere often found birds to be at greatest risk of harm. Consequently, the SFMSP began sponsoring a series of field and laboratory studies and experiments to define the potential risk of mercury to wading birds.

The work is ongoing and, as stated in the “Background” section, there is, as yet, no clear evidence that wading bird populations are harmed or limited by mercury. Because wading birds have been identified as one of the key success indicators of the Comprehensive Everglades Restoration Plan (CERP, Chapter 10), interest in wading bird ecology and population health has continued unabated. In previous years, District scientists and scientists working on behalf of the Sugar Cane Growers Cooperative of Florida have performed in-depth and progressively more sophisticated formal toxicological risk assessments of mercury risks to Everglades wading birds. These studies were treated extensively in the initial report of this series (*Everglades Interim Report*, Chapter 7, pp. 40-46, SFWMD, 1999) and in the *2000 Everglades Consolidated Report* (2000 ECR, pp. 21-29, SFWMD, 2000). These analyses are not repeated here. For a thorough treatment of ecological risk assessment analyses, refer to Chapter 7 of the documents referenced above.

Though no new, formal ecological risk assessment is presented in this document, field monitoring and research continue to refine knowledge of mercury biogeochemistry, bioavailability, and food web biomagnification. Such information is key to developing an improved understanding of the food web relationships among mercury in prey and predator, and to supporting further refinements in estimation of ecological risk.

RESPONSE OF THE NATURAL SYSTEM TO SOURCE REDUCTIONS

Small numbers of largemouth bass collected at three Everglades locations (L-38A, L-35B and L-67A) in 1988 and reported in early 1989 averaged nearly 2.5 mg/kg (ppm) total mercury in the edible fillet. These findings were promptly confirmed and led to the unprecedented issuance of health advisories to fishermen by the Florida Department of Health to cease consumption of largemouth bass from that area. Subsequent sampling indicated that mercury problems extended to many other Florida waters. From that time, to determine whether the trend of mercury in fish is increasing or decreasing, the FWC, the Department of Health, and the FDEP began collaborating on annual collection and testing of fish from five sites in Florida, including the L-67 site.

Subsequent monitoring of mercury in fish and wildlife in the Everglades and elsewhere in Florida has yielded annual information on mercury body burdens in nestlings that can be similarly examined for time trends in an update of the corresponding figure from the 2001 ECR, showing a continuing, if small, decline in mercury in largemouth bass (**Figure 2B-14**). **Figure 2B-15** shows a similar time course of mercury levels in the feathers of great egret nestlings.

How these trends in mercury in biota compare to trends in mercury load to the Everglades (**Figure 2B-7**) or to emissions trends in the United States or Florida has been difficult to determine because of a lack of data, models, and other tools to model the local, regional and global scales of air-pollutant cycling.

Atmospheric deposition trend monitoring of rainfall mercury deposition began in South Florida with the establishment of four monitoring sites of the Florida Atmospheric Mercury Study (FAMS) adjacent to the Everglades in 1994 and 1995 and continuing through 1996. In 1995 the FDEP sponsored the installation of one of the first Mercury Deposition Network (MDN, a sub-network of the National Atmospheric Deposition Program) sites at the ENP Beard Center, collocated with the FAMS site. The sites, operated by their respective groups side-by-side for 15 months, established that comparability was excellent. After completion of the FAMS project, the District assumed responsibility for the ENP MDN site, and also established two others (Andytown and ENRP) to ensure continuity of long-term trend monitoring of atmospheric mercury wet deposition to the Everglades. Recent meta-analysis of mercury wet deposition from both FAMS and MDN does not indicate any significant trend (Pollman and Atkeson, 2001, in prep). It is likely that emissions reductions occurred before the monitoring began in 1994. It is also likely that data variability will hamper trend detection in deposition data.

Table 2B-2. Summary of THg concentrations (mg/Kg wet) reported for liver and tail muscle from Florida alligators (Rumbold et al., in press)

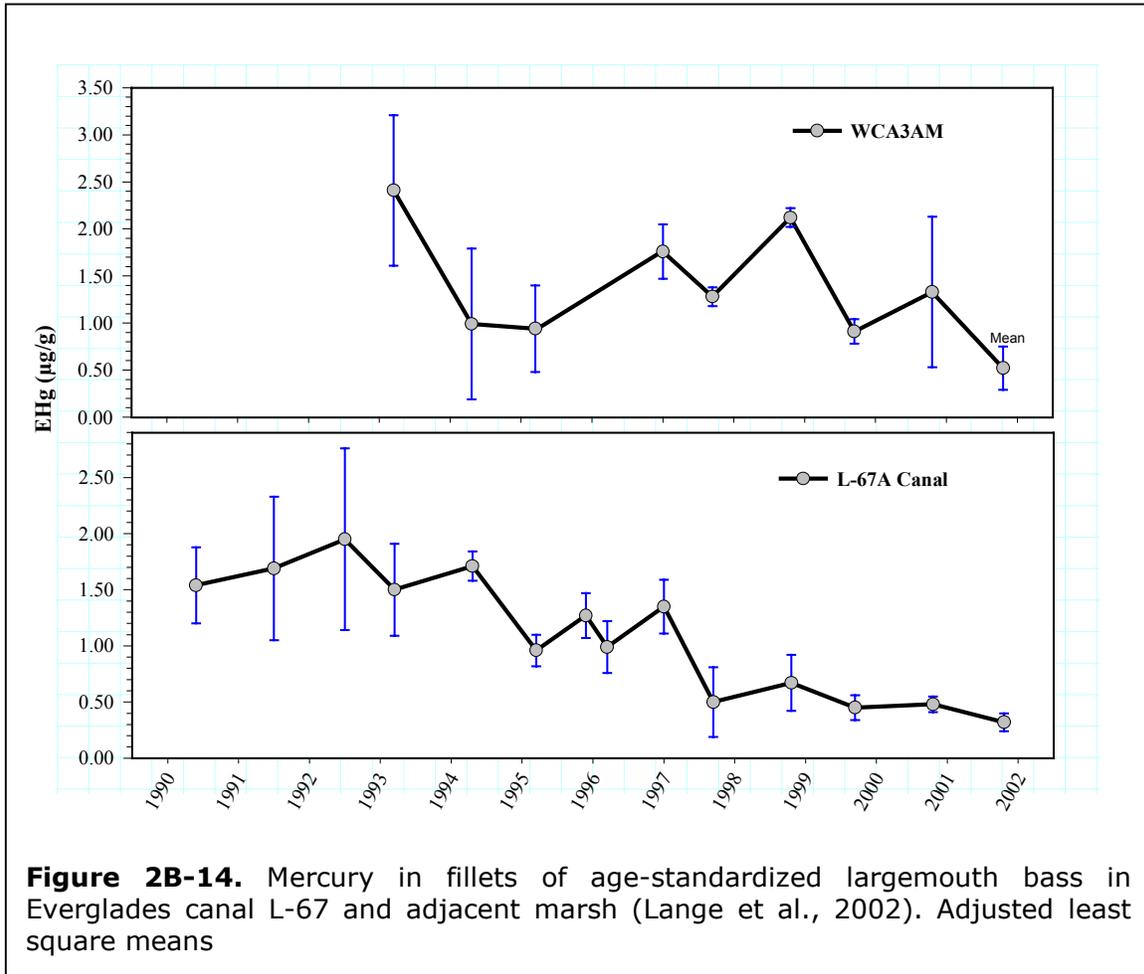
Collection Location	Collection Year	Alligator Size (cm)	n	Liver THg (mg/Kg)	Muscle THg (mg/Kg)	Source
North Florida						
N. Fl. lakes	1985	290-380	32	NR	0.305 ±0.21 ^a	Delany et al. 1988
N. Fl. lakes	1997	36-40	31	0.4 ±0.08	0.06 ±0.02	Burger et al. 2000
SOUTH						
Everglades	1989	NR	NR	NR	2.9 ^b	Ware et al. 1990
WCA-2 and 3	1989	120-280	18	NR	2.36 ±0.99	Hord et al. 1990
Shark River Slough	< 1991	NR	5	NR	2.96 ^c ±1.2 ^a	Roelke et al. 1991
WCA-2 and 3	1992-93	200-350	12	39.99 ±24.05	2.61 ±0.91	Heaton-Jones et al. 1997
Lake Okeechobee	1992-94	162-221	12	NR	0.23 ±nr	Unpubl. FWC data
WCA-3A north	1992-94	124-193	11	NR	1.62 ±nr	Unpubl. FWC data
WCA-3A south	1992-94	122-289	11	NR	1.89 ±nr	Unpubl. FWC data
WCA-3	1994	121-196	18	10.2 ±6.22 ^d	1.17 ±0.36 ^d	Jagoe et al. 1998
WCA-1, 2, 3, ENP and BCNP	1999	118-185	28	4.89 ±3.99	0.64 ±0.4	This study

NR = not reported

- a. Standard error
- b. Size and exact locations not reported; value scaled off graph
- c. Geometric mean
- d. Converted from dry-weight basis using reported moisture content

Data from about 1994 to the present suggest mercury levels are declining in Everglades fish and birds. This apparent trend is consistent with the timing and extent of a national trend in the mercury content of incinerated trash in the United States. While this evidence is preliminary, it is consistent with the time lag predicted by modeling for a decline in atmospheric deposition

resulting from decreasing amounts of mercury emitted by air sources within South Florida. Further declines in wildlife mercury exposure from these control measures are possible. Additional controls are possible and could produce greater reduction in exposure. With existing evidence, it is premature to rule out the possibility that emissions controls can further reduce exposures in the entire Everglades, including the impacted areas.



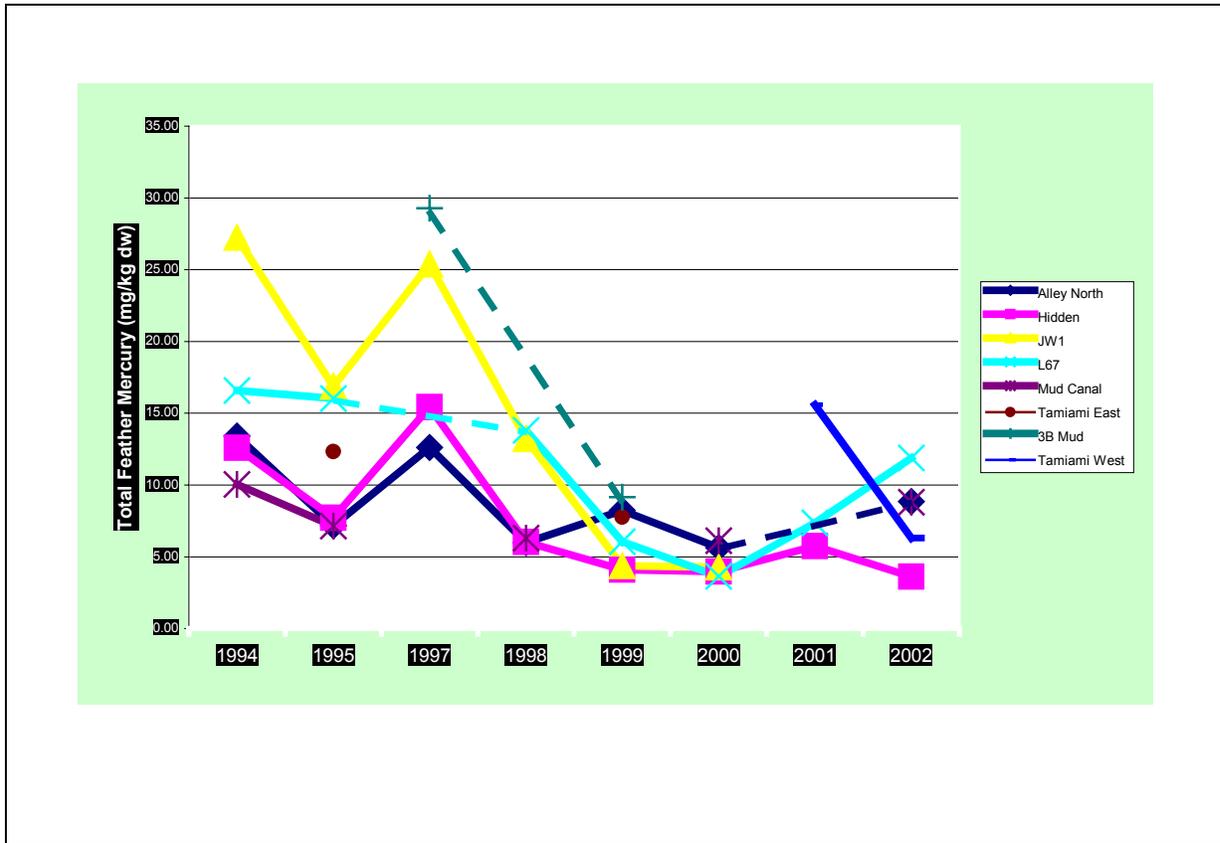


Figure 2B-15. Mercury concentration in the feathers of great egret nestlings standardized to 8-cm bill length (Frederick and Spalding, 2002)

Because the data are limited, this continuing trend remains a working hypothesis that will be subject to data evaluation from other sites within the Everglades and Florida and to rigorous statistical analysis. Monitoring of mercury trends in atmospheric deposition, fish and wading birds will continue indefinitely. It is likely that much of the emissions reduction responsible for this apparent trend occurred prior to the initiation of mercury monitoring in wet deposition in South Florida. The time lag between emissions reduction and fish and bird reduction is consistent with the lag predicted by E-MCM modeling. Further work consisting of hind-casting emissions and examination of new sediment cores is underway to test this hypothesis.

REGULATORY IMPLICATIONS

ADDITIONAL EMISSIONS CONTROLS

Mercury affecting the Everglades is deposited from the atmosphere directly onto the Everglades and on the watershed of tributary inflows, which passes some mercury into the Everglades. By reducing atmospheric deposition rates through control of emission sources, mercury impacts can be reduced. Sources of atmospheric mercury emissions are both natural and man-made and may be defined as local (i.e., sources within 100 km of the Everglades), regional (i.e., within 1,000 km of the Everglades) and global. These three spatial scales roughly

correspond to sources under Florida's regulatory control, those that may be regulated by the USEPA, and those that can be addressed only by international agreement. Mercury emissions in South Florida have declined significantly in the past 10 years. Mercury in municipal solid waste began declining in the late 1980s because of removal of mercury from consumer and industrial products (USEPA, 1992). Following the implementation by the FDEP of the first mercury-limiting standard for municipal waste combustors in 1994, mercury emissions have continued to decline. In addition, USEPA regulation of medical waste incinerators since the early 1990s rendered small incinerators uneconomic, and virtually all have ceased operation; mercury emissions from this sector have declined from 8,815 lb (14 percent) in 1990 to 467 lb (3 percent) in 1996. Combined mercury emissions from both sources have declined by approximately 95 percent over the same period. The FDEP has vigorously pursued other mercury reduction strategies, from mandating the recycling of mercury-containing lamps and devices, to pollution prevention initiatives and working with the health care industry to reduce mercury in the chronically high hospital waste stream. The effect of these emissions reduction measures will not be fully realized for years, as previously deposited mercury is slowly bound up in sediments, is evaded, or is otherwise rendered less bioavailable.

Before undertaking further steps to control mercury emissions from local sources, it is essential to know the benefits, expressed as the decline in total mercury deposition. Computing the decline in total mercury deposition that could result from FDEP regulation of local sources requires numerical values for the total mercury deposited on the Everglades from both local and background sources, the fraction contributed by local sources and the efficacy of control technologies as applied to local sources. These can be estimated by application of the data and models described above. Reduction in the rate of atmospheric deposition of mercury would reduce levels of toxic methylmercury in top predators. Studies performed with the Everglades Mercury Cycling Model show the response will be nearly linear, i.e., a 25 percent reduction in deposition will result in almost a 25-percent reduction in accumulation of methylmercury in top predators. The initial decrease in predator mercury levels will be rapid, within a decade or so, and the response to reduced mercury load will be essentially complete within two to three decades.

CONCLUSIONS

ADEQUACY OF EXISTING MANAGEMENT STRATEGIES

The public and private agencies comprising the SFMSP have effectively worked to accomplish the following:

1. Describe and define the mercury problem in the Florida Everglades
2. Identify and quantify the sources and causes of the mercury problem
3. Develop and implement appropriate environmental controls to abate the mercury problem and monitor the effectiveness of the abatement measures

A comprehensive program of monitoring, modeling and research has broadened the understanding of the sources and causes of the mercury problem. The results have been incorporated into sophisticated environmental models that predict the Everglades will respond to decreases in atmospheric mercury deposited into the marshes in a direct, nearly one-to-one relationship. More encouragingly, the model suggests that significant benefits from decreased mercury loading should be seen in less than a decade, with full benefits being achieved within a generation. Current monitoring trends in mercury within the Everglades system indicate the

beginnings of positive results from the pollution prevention and control efforts that began in the mid-1990s.

As a result of a series of international, North American and Florida initiatives, mercury usage in North America has declined approximately 90 percent since 1990. In addition, environmental controls have been developed and implemented for medical waste incineration (MWI) and municipal solid waste incineration, both of which have resulted in emissions declines in excess of 95 percent for each source sector. The major pending decision with regard to control policy lies with the USEPA, which, having made its decision under the Clean Air Act Amendments of 1990 to regulate mercury emissions from coal- and oil-fired utility boilers, must develop regulatory specifics by the end of 2003. When the USEPA regulations are promulgated, Florida, as one of the delegated states, will be responsible for local implementation.

The atmospheric mercury studies conducted by the FDEP and its collaborators in the SFMSP have been conducted in close collaboration with the USEPA to ensure rapid transfer of new technologies or information into the national program. The FDEP is proud that it has been able to provide useful information to the USEPA and other federal agencies, and has thereby assisted with and promoted a sound scientific basis for policy decisions.

The multi-agency approach to the mercury problem in South Florida has been a notable example of the successful marriage of science and policy. This comprehensive, long-term approach has enabled Florida to become the model for addressing a complex, multimedia environmental problem.

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